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Risø-R-437
Part I (Main text)

RISØ

Risø-R-437
Part I (Main text)

Environmental Studies on Radioecological Sensitivity and Variability with Special Emphasis on the Fallout Nuclides ^{90}Sr and ^{137}Cs

DK 8100066

Asker Aarkrog

Risø National Laboratory, DK-4000 Roskilde, Denmark
June 1979

ENVIRONMENTAL STUDIES ON RADIOECOLOGICAL SENSITIVITY AND VARIABILITY WITH SPECIAL EMPHASIS ON THE FALLOUT NUCLIDES ^{90}Sr and ^{137}Cs

Asker Aarkrog

Denne afhandling er af det naturvidenskabelige fakultet ved Københavns Universitet antaget til offentligt at forsvares for den naturvidenskabelige doktorgrad.

København, den 25. november 1980 Claus Nielsen
dekan

Forsvaret vil finde sted fredag d. 27. marts 1981 kl. 14
i Annex auditorium A, Studiestræde 6, København K.

RISØ-R-437

Part one
Main text

ENVIRONMENTAL STUDIES ON RADIOECOLOGICAL SENSITIVITY AND VARIABILITY WITH SPECIAL EMPHASIS ON THE FALLOUT NUCLIDES ^{90}Sr and ^{137}Cs

Asker Aarkrog
Health Physics Department

Abstract. Radioecological sensitivity and variability are quantities that are used to characterize the radioecological properties of environmental samples.

The radioecological sensitivity is the infinite time-integrated radionuclide concentration in the environmental sample considered arising from a deposition of 1 mCi km^{-2} of the radionuclide in question.

This quantity makes it possible to compare various environments as to their vulnerability to a given radioactive contamination.

The variability of the concentrations of a radionuclide in an environmental sample, with respect to a given parameter, is defined as the partial coefficient of variation due to this parameter. (Continued next page)

June 1979
Risø National Laboratory, DK 4000 Roskilde, Denmark

The variability with time is a useful way to assess the route of contamination of the sample and the local variability is a measure of environmental inhomogenity with respect to radioactive contamination.

Radioecological sensitivity and variability were applied to the ^{90}Sr and ^{137}Cs data obtained from the environmental studies on the human foodchain carried out during the last two decades in Denmark, the Faroe Islands, and Greenland.

The per caput effective dose-equivalent commitments from radioactive debris from nuclear weapons testing was estimated to be 1.6 mSv in Denmark, 4.2 mSv in the Faroe Islands, and 1.6 mSv in Greenland.

INIS-descriptors: AIR, ATMOSPHERIC PRECIPITATIONS, BODY, BREAD, CESIUM 137, CEREALS, COMPILED DATA, CONTAMINATION, DENMARK, DIET, DOSE COMMITMENTS, DRINKING WATER, ENVIRONMENT, ENVIRONMENTAL EXPOSURE PATHWAY, EXPERIMENTAL DATA, FAROE ISLANDS, FISHES, FOOD CHAINS, FRESH WATER, FUCUS, GRAMINEAE, GLOBAL FALLOUT, GREENLAND, LICHENS, MAN, MEAT, MILK, RADIOACTIVITY, RADIOECOLOGY, RADIOECOLOGICAL CONCENTRATION, RADIONUCLIDE KINETICS, RADIONUCLIDE MIGRATION, SAMPLING, SEAWATER, SEDIMENTS, SEEDS, SENSITIVITY, SOILS, STATISTICAL MODELS, STRONTIUM 90, TEETH, TRANSLOCATION, VEGETABLES.

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To my collaborators

Nu gik da Mange til Skoven for det gode Lønnebrød; Skyld, men der var kun Een, som kom hjem med et Slags Forklaring; Ingen havde vovet dybt nok inde, og han da ikke heller, men han sagde dog at Klokke-Lyden kom fra en meget stor Ugle i et hult Træ; det var saadan en Visdoms-Ugle, som idelig slog sit Hoved mod Træet, men om Lyden kom fra dens Hoved eller fra den hule Stamme, det kunde han ikke endnu med Bestemthed sige, og saa blev han ansat som Verdens Klokker og skrev hvert Aar en lille Afhandling om Uglen; men ligemeget vidste man.

KLOKKEN

Numbers of people now went to the wood in the hope of getting such a good appointment, but there was only one who came home with any kind of explanation. None of them had been far enough into the wood, nor had this man either, but all the same he made out that the bell-sound came from a very big owl in a hollow tree. It was a kind of owl of wisdom that kept knocking its head against the tree; but whether the sound came from the owl's head or from the hollow trunk he couldn't yet say with any certainty. So he was appointed Universal Bell-ringer, and every year he wrote a little essay on the owl, but no one was any wiser than before.

THE BELL

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GENERAL INTRODUCTION

Shortages of natural resources and increasing pollution have in recent decades made an understanding of the interaction between man and nature even more necessary. As compared to most chemical pollution, the radioactive contamination of the environment is modest. The prospects of increasing nuclear power production in the next decades, however, justify radio-ecological studies. Radioactive contamination of the human environment became a reality on 16 July 1945, when the first fission weapon was tested near the town of Alamogordo in New Mexico. Military use of nuclear energy is still the main source of global radioactive contamination; atmospheric test explosions with thermonuclear weapons such as those still performed by the People's Republic of China thus contribute measurably to the concentrations of radionuclides in man's environment, even far distant from the test sites. Accidents involving nuclear weapons, e.g., those that took place at Palomares, Spain, and at Thule, Greenland, have resulted in local contamination, but were not of global concern with respect to pollution. The use of nuclear energy to produce power has also given rise to measurable concentrations of radionuclides in the environment, especially close to nuclear installations; however, the global mean dose rate originating from nuclear energy production is at present (1979) nearly two orders of magnitude less than that from military applications.

Radioecology comprises the relationships between radioactive substances or radiation and the environment. Thus this science includes the movement of radionuclides within ecological systems and their accumulation within specific ecosystem components such as air, water, soil and living organisms. The effects of ionizing radiation upon ecological systems, which are not dealt with in this study, may best be termed "radiation ecology", although this last term is used by some authors (Sc72) instead of "radioecology".

The movement and accumulation of radionuclides in the environment may be studied in two different ways. Radionuclides may be experimentally introduced in known amounts into a limited and well defined environment, or use may be made of the contamination already present in the natural environment. Each method has its merits and limitations. The experimental method may shed light on single mechanisms acting in the environment, but it is not able to describe the whole complex of interactions and combinations that makes up natural conditions. The second method, the environmental approach, yields information on the variation of radioactive contamination in the environment studied, but it gives little information on the various processes responsible for the levels observed. Neither of the two methods is thus ideal, but one complements the other. In the present study, emphasis has been laid upon the last method.

A radioecological study of environmental samples may have several aims. A primary purpose is often to ensure that certain limits of contamination are not exceeded, in other words to ensure the radiological protection of man. A further purpose may be to set up models based on the data collected, and then from these models to predict the levels resulting from known releases of a contaminant to the environment and from the predicted radionuclide levels to assess the doses to man from the release. Finally, we may intend to make a study of various processes in the environment, in which case radioactive substances are used as tracers.

The present study intends to identify and define quantities that characterize the radioecological properties of environmental samples. Radioecological sensitivity and variability are the two quantities used for this purpose.

The radioecological sensitivity of a sample is the infinite time integral of appropriate quantities of the sample from an appropriate quantity of the radionuclide deposited (cf. C.3). The radioecological sensitivity equals the steady state concentration in the sample of the radionuclide considered from a constant annual deposition rate of the radionuclide distributed like global fallout throughout the year. Radioecological sensi-

tivity makes possible a comparison between various environments of the vulnerability to a given radioactive contamination. Such information is useful for the planning to be made in connection with releases of radionuclides from nuclear installations.

The variability of radionuclide concentrations in an environmental sample with respect to a given parameter is the partial coefficient of variation due to this parameter (cf. B.4), determined by means of an analysis of variance. The variability gives indirect information on the mechanisms of contamination for the various samples. It is useful as an indication of the necessary number of samples and their frequency in environmental monitoring programmes for nuclear facilities. Furthermore, the variability may contribute to an assessment of the maximum concentrations of a contaminant likely to occur in a given environment.

The present study began as a pre-operational study in 1956 around the Risø site. Later, when the research establishment came into operation, the measurements became the operational surveillance of the environmental radioactivity at Risø. In co-operation with the Danish National Health Service, the measurements were extended in 1959 to a general study of radioactive contamination in Denmark. Three years later the Faroe Islands and Greenland were also included in the studies. Contamination at that time was the result of the atmospheric nuclear tests carried out by the USSR, the USA and the UK.

Since 1957 the results of these radioactivity measurements have been published in a series of Risø Reports (RRD59-76, RRF62-76, RRG62-76), which contain the basic data for this study. The data were compiled in the so-called STATDATA program (Li75), which makes feasible the treatment of the approx. 40,000 data.

The present dissertation is divided into two parts: the main text, and four appendices. The main text comprises five chapters. The first chapter deals with the abiotic environment: air, water and soil, which are the primary recipients of radioactive pollution. Chapter two comprises the radioactive contamination of the producers, with emphasis on cereal grain, which is a main

contributor to the radionuclide content of the Danish diet. The radioactive contamination of the consumers and their products, especially milk, are treated in the third chapter. Chapter four discusses the contamination of the human diet and tissues. The conclusion to chapter four furthermore contains an estimate of the population doses due to the contamination from nuclear weapons testing. Chapter five is a general discussion of important features of the four preceeding chapters.

Details of methods and procedures are contained in the appendices. Appendix A summarizes sampling programmes, radiochemical procedures, the various methods used for the measurements of the ionizing radiation emitted by the samples, and the estimation of errors. Appendix B contains the statistical procedures mentioned and tables with analysis of variance (anova) and the estimates of the variability of the various samples. The prediction models used for the estimation of the radioecological sensitivity are shown in appendix C, while appendix D contains various notes to the main text.

As ^{90}Sr and ^{137}Cs are the two fallout nuclides that contribute most to the dose from fallout to present generations, this study puts emphasis on these two nuclides. Not all sample items, areas and periods have been covered with the same intensity and thoroughness. Importance has been attached to those samples assumed to be the most pertinent. The samples were obtained from Denmark, the Faroe Islands and Greenland. Environmental conditions (meteorology, soil characteristics) as well as the habits of life show marked differences in these three areas; and studies of the radioecological sensitivity and variability of environmental samples from Denmark, the Faroe Islands and Greenland may give an idea of the variation to be expected of these quantities within the north Atlantic region.

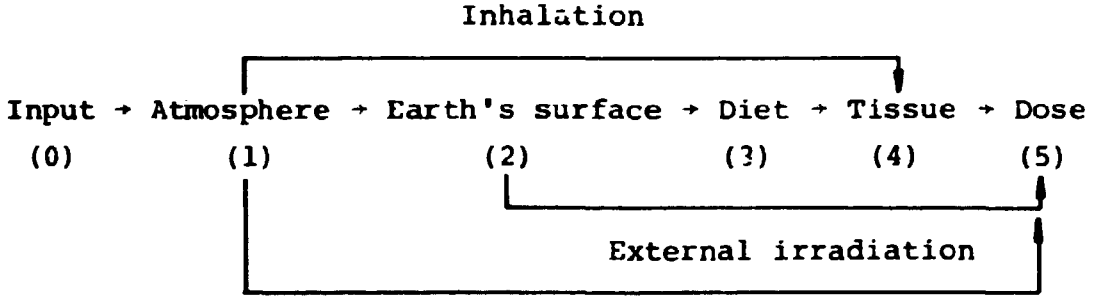
1. AIR, WATER AND SOIL (the abiotic environment)

1.1. Introduction

The atmosphere is the primary recipient of airborne radioactive contaminants, whether these pollutants originate from atmospheric nuclear test explosions or from airborne releases from nuclear installations. The air is the medium in which the contamination is dissipated and transported.

Radioactive contamination of the atmosphere may result in doses to living organisms from three exposure routes: by direct radiation from the debris suspended in the air, from inhalation (assimilation), or indirectly from contamination of the food chains.

UNSCEAR (Un77) applies the model:



Nuclear debris may be deposited directly from the atmosphere on the surface of the earth, as so-called dry fallout; but a substantial amount of the radioactive contamination in the air reaches the earth with precipitation. The term direct contamination of crops is used when the above-ground parts of crops collect airborne debris, or adsorb radionuclides from precipitation.

In the case of water-borne releases, e.g. waste-water from nuclear installations, the sea or fresh-water systems are the primary recipients of the contaminants. In analogy with air, the doses from water-borne radioactive contaminants are delivered either by external radiation from the activity in the

water, or from drinking (assimilation) of the water, or indirectly from contamination of aquatic or (in the case of irrigation) terrestrial food chains, respectively.

Soil is normally not a primary recipient of unsealed radioactive contaminants, but underground nuclear explosions provide an exception. Soil usually receives contamination from air or from water. The transport of radioactive substances in the soil is in general connected to water transport. Under dry conditions especially, contaminated soil may be resuspended in the air by the wind. The activity in the soil strongly influences the indirect contamination of crops, i.e. contamination due to root uptake. In the special case of resuspension there may, however, be direct contamination from the soil.

1.2. Air

Naar Vinden løber hen over Græsset, da kruser det sig som et Vand, løber den hen over Kornet, da bølger det som en Sø, det er Vindens Dands;
VINDEN FORTÆLLER OM VALDEMAR DAAE OG HANS DØTRE

When the wind sweeps over the grass, it ruffles it like water; and when it sweeps over the corn, it goes surging like the sea. That's the dance of the wind.
THE WIND TELLS THE STORY OF VALDEMAR DAA AND HIS DAUGHTERS

In atmospheric thermonuclear weapons testing (cf. D.1.2.), which has been the main contributor to anthropogenic radioactive contamination with long-lived radionuclides, nearly all the debris (∿ 99%) is injected into the stratosphere. From here the fallout descends mainly in the spring through the tropopause into the troposphere with a mean residence time in temperate latitudes of the order of 1 year. The residence time for particulate radionuclides in the troposphere is about one month. The debris, which has a particle size of the order of 0.1-1μ (Pei65) is removed from the troposphere by three processes: rain-out caused by droplet formation within clouds, washout by falling raindrops picking up radionuclides, and dry deposition on land surfaces or plant cover (Un77).

The first systematic studies of important fallout nuclides in air were initiated when thermonuclear test-explosions began. In the U.K., air samples have thus been collected since 1952 at Chilton (Pei60) and analyzed for ^{137}Cs. A world-wide study

of radionuclides in surface air was started in 1957 by the U.S. Naval Research Laboratory; since 1963 this study has been continued by HASL (the Health and Safety Laboratory of the U.S. Atomic Energy Commission) (Ha58-78).

In Denmark, the National Defence Research Council (Am58) initiated fallout studies of air and precipitation in the mid fifties. Systematic investigations of radiostrontium in air samples collected at Risø began in 1957 (RRD58-59).

1.2.1. Strontium-90

The time variation of fallout nuclide concentrations in air is mainly controlled by the number and intensity of atmospheric nuclear test-explosions and by the meteorology of the earth's atmosphere. The first factor is chiefly responsible for the variation among years, while the second primarily influences the seasonal variation.

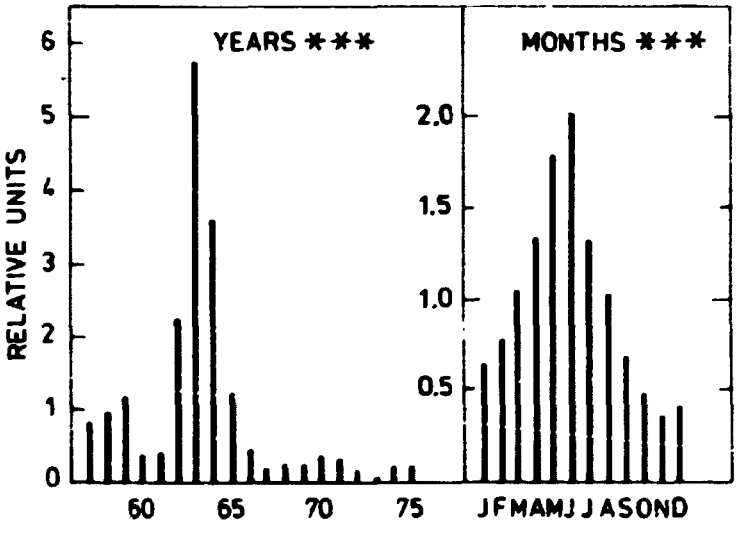


Fig. 1.2.1. The annual and monthly variation of ^{90}Sr in ground-level air collected at Risø 1957-1975. The bars indicate the air concentrations relative to the grand mean: 5.31 fCi m^{-3} (= 1 at the relative scales).

An anova (analysis of variance) (table B.1.2.1.) of ^{90}Sr in ground level air collected at Risø showed highly significant variations among years and among months. The maximum appeared in 1963 after the 1962 test series (cf.D.1.2.) As shown in fig.

1.2.1, the maximum within the year occurred in May-June and the minimum in November-December; although the anova showed an interaction between years and months, all years displayed the so-called spring peak. The variability among years (table B.1.2.3.) was 1.45 for the period 1957-1975, and among months it was 0.58.

As the air samples in the present material were collected at one location only, there is no direct means of studying a possible local variation. The air sampling programmes conducted by HASL (Ha58-78) and by Harwell (Ae58-74) may, however, (cf.D.1.2.1.) be applied for an assessment of the local variation in Denmark, which was estimated to be negligible.

The integrated air activity concentration for the entire period of nuclear testing (up to 1975) was estimated from table D.1.2.1. at 121 fCi $^{90}Sr\ m^{-3}$ y. This quantity arose from 12.13 MCi ^{90}Sr deposited in the northern hemisphere (Un77), (local fallout at the test sites excluded). Hence, 1 MCi ^{90}Sr deposited in the northern hemisphere corresponded to 10 fCi $^{90}Sr\ m^{-3}$ y in ground-level air collected in Denmark. However, the infinite time integral of the air activity may also be related to the total amount of ^{90}Sr dissipated as global fallout, i.e., 16 MCi. In which case, 1 MCi ^{90}Sr results in 7.6 fCi $^{90}Sr\ m^{-3}$ y in ground-level air in Denmark. As the majority of tests were carried out in the northern hemisphere, the figure was considered representative of injections in the northern hemisphere.

The half residence time of ^{90}Sr has been earlier (X) estimated at 10 months for the period 1963-1967; the inclusion of the post-test periods 1959-1961 and 1971-1973 confirmed this estimate.

1.2.2. Cesium-137

According to UNSCEAR (Un77), the ratio between ^{137}Cs and ^{90}Sr in nuclear weapons debris is 1.6. This ratio is higher than the ratio at formation, which according to HARLEY is 1.45 (Harl72). It is not clear whether this difference is due to analytical errors or to fractionation phenomena (Sh75). If the

$^{137}Cs/^{90}Sr$ ratios in the Risø air samples are considered, the anova shows no significant seasonal variation. The variation between the years was significant; however, the time variation observed for the Risø filters did not reflect a general global variation of the $^{137}Cs/^{90}Sr$ ratio in air (cf.D.1.2.1.). Hence it was, as earlier suggested (X), assumed that the $^{137}Cs/^{90}Sr$ ratio in airborne debris at an annual base was nearly constant in time and equal to 1.6.

The variability of the ^{137}Cs air concentrations among years was 1.65 (1957-75) and among months 0.64, i.e. a little higher but not significantly different from that of ^{90}Sr. If ^{90}Sr and ^{137}Cs are considered together during the period 1962-1975, when precipitation data are available too, the variability among years becomes 1.72 (table B.1.2.3.).

From table D.1.2.1. the integrated air activity for the entire period of nuclear testing (up to 1975) was estimated at 222 fCi $^{137}Cs\ m^{-3}$ y. This estimate is comparable to that obtained from Chilton in the U.K. (Ae58-74) for the period 1954-1975 ($\sim$ 240 fCi $^{137}Cs\ m^{-2}$ y). The total production of ^{137}Cs from nuclear weapons testing was 1.6 • 16 MCi = 25.6 MCi, hence the transfer factor from input ground-level air was 8.7 fCi $^{137}Cs\ m^{-3}$ y per MCi ^{137}Cs injected. This is 15% higher than the corresponding factor for ^{90}Sr. As the half-lives of the two nuclides are nearly the same, the infinite time integrals of the air concentrations per MCi were expected to be nearly identical. The difference was mainly due to the measured ratio of $^{137}Cs/^{90}Sr$ in the air samples being higher than the adapted ratio of 1.6.

1.2.3. Strontium-89

Strontium-89 is detectable in fresh fallout together with ^{90}Sr; but, due to its relatively short half-life (50.5 days), ^{89}Sr is seldom measurable more than a year after its creation. As the $^{89}Sr/^{90}Sr$ ratio at formation is 185, according to UNSCEAR (Un77), this ratio decays to approx. 1 in one year. Strontium-89 is thus often difficult to determine because the measurement takes place on the background of the ^{90}Sr activity (A.2.1.). According to HASL (Har172), the $^{89}Sr/^{90}Sr$ ratio at formation is

147, but Risø's measurements of the ratio based on precipitation and air samples have indicated that the UNSCEAR value of 185 is the best fit to the observed data (RRD63).

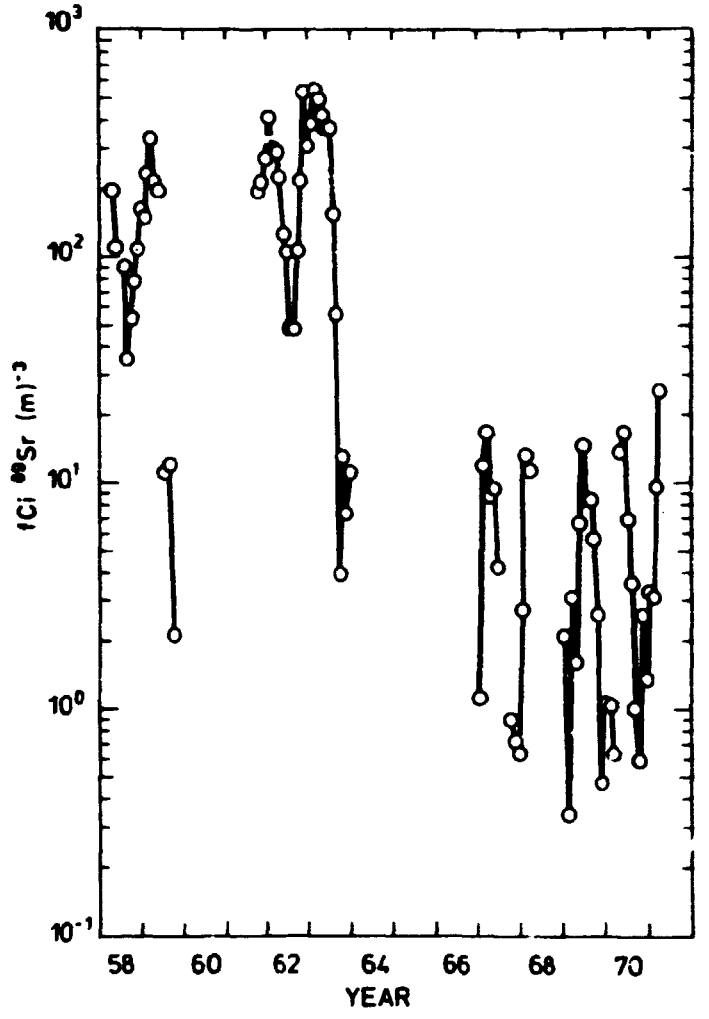


Fig. 1.2.3. Strontium-89 concentrations in ground-level air collected at Risø 1958-1971.

Figure 1.2.3. shows the ^{89}Sr levels in monthly air samples collected since 1958. The data were complete for the period 1961-1963; the ^{89}Sr air debris in this period was created in the 1961-1962 test series when approx. 9 MCi ^{90}Sr (estimated from UNSCEAR (Un77)) or 1665 MCi ^{89}Sr were injected into the northern hemisphere. The integral air level during this period was 514.5 fCi ^{89}Sr y m^{-3}. Hence, 1 MCi ^{89}Sr corresponded to 0.3 fCi ^{89}Sr m^{-3} y. During the periods 1958-1959 and 1967-1971, the data were incomplete; during these periods 1 MCi ^{89}Sr yielded approx. 0.4 and 0.2 fCi ^{89}Sr m^{-3} y, respectively. An integral air level of 0.3 fCi ^{89}Sr m^{-3} per MCi ^{89}Sr was a reasonable average, and for the entire period of nuclear testing the integral air level becomes: 16 · 185 ·0.3 = 890 fCi ^{89}Sr y m^{-3}.

1.2.4. Manganese-54

During the intensive test series that took place in the USSR in 1961, ^{54}Mn, (Gu64) which is a neutron activation product, was produced in such quantities that it was measurable in ground-level air samples collected during 1962-1966 (fig. 1.2.4.). The integral level was 149 fCi ^{54}Mn y m^{-3}. This is in agreement with the mean level: 132 fCi ^{54}Mn y m^{-3} found at the four U.K. stations: Chilton, Milford Haven, Orfordness and Eskdalemuir (51^{o} - 55^{o} N) (Ae58-74), but incompatible with the UNSCEAR figure (Un77) of 1130 fCi ^{54}Mn y m^{-3} based on HASL measurements (Ha58-78) from Sterling in Virginia (39^{o} N). If the Sterling air levels are compared with the rainwater concentrations from Westwood in New Jersey (41^{o} N) from 1963-1965, (Ha58-78) it is noticed that the washout ratio (cf. 1.3.2.) is 0.06 $\pm$ 0.02 (1 SE), while the washout ratio usually found is an order of magnitude larger (En71). It is therefore concluded that the Sterling ^{54}Mn air data used by UNSCEAR were probably an order of magnitude too large.

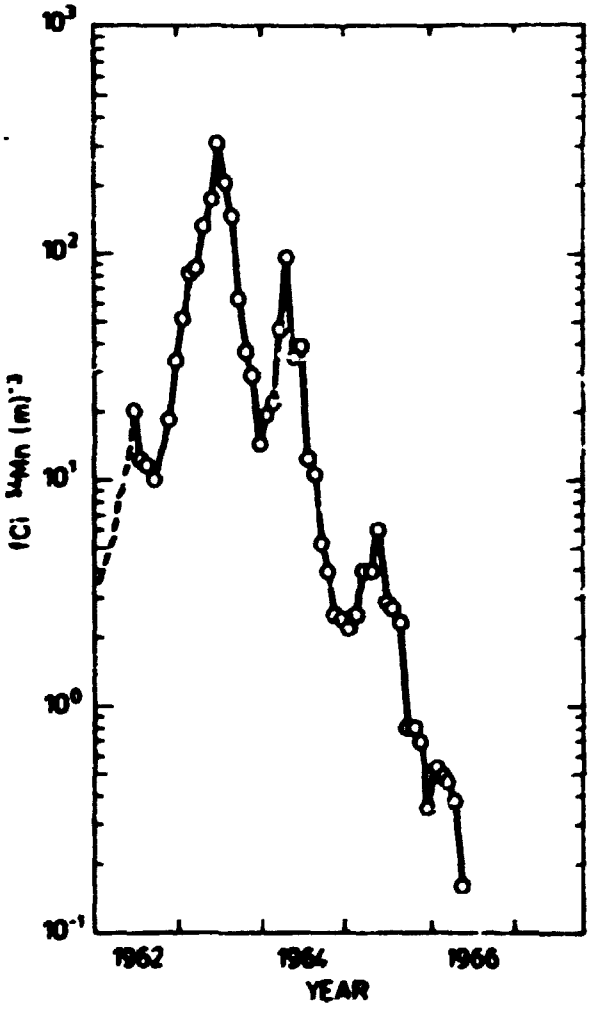


Fig. 1.2.4. Manganese-54 concentrations in ground-level air collected at Risø 1962-66.

The effective half-life of ^{54}Mn in the atmosphere was approx. 5 months, which corresponds to the theoretical value considering that the half residence time of long-lived debris was 10 months. The total production of ^{54}Mn in the 1961-1962 test series was estimated at 54 MCi (cf. table 1.2.5.).

1.2.5. Other radionuclides

Throughout the years a number of other radionuclides have been studied in ground-level air at Risø.

Cerium-144 has been measured regularly in most years. From the test series in 1961-1962, approx. 430 MCi ^{144}Ce yielded an integral air level of 1278 fCi ^{144}Ce y m^{-3}, or 1 MCi ^{144}Ce corresponding to 3.0 fCi ^{144}Ce y m^{-3}. From this ratio the total integral air level at Risø from 762 MCi ^{144}Ce (cf. table 1.2.5.) was estimated at 2.3 pCi ^{144}Ce y m^{-3}. This estimate is comparable to those for Moonsonee (51° N) of 2.5 pCi ^{144}Ce y m^{-3} (Ha58-78) and for Chilton (51° N) of 2.0 pCi ^{144}Ce y m^{-3} (Ae58-74). The effective half-life of ^{144}Ce in air samples collected in the years 1963-1966 was approx. 5 months, i.e. equal to the observed effective half-life of ^{54}Mn in air.

Europium-155 was measured in air filters collected from the middle of 1961 to the end of 1966 (III). The integral air level for this period was 26 fCi ^{155}Eu y m^{-3}. The production of ^{90}Sr available as global fallout was approx. 9 MCi in the 1961-1962 test series*), and as the ratio $^{155}Eu/^{90}Sr$ at formation was estimated at 0.66 (III), the globally available ^{155}Eu became 6 MCi; 1 MCi ^{155}Eu thus corresponded to 4.4 fCi ^{155}Eu y m^{-3}.

During 1971-1974 ^{95}Zr was determined in air samples. The air activity in this period resulted from atmospheric test series in 1970-1973, and was equivalent to 0.4 MCi ^{90}Sr (table D.1.2.1.) or 90 MCi ^{95}Zr (table 1.2.5.). The observed integral air level was 40 fCi ^{95}Zr m^{-3} y, hence 1 MCi ^{95}Zr injected into the northern hemisphere resulted in 0.44 fCi ^{95}Zr m^{-3} y.

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*) This ^{90}Sr yield was lower than the previous estimate (III) which was based on an early yield estimate (Fe63) of these test series

If the transfer factors: T (fCi m^{-3} y per MCi) are plotted against the effective half-lives in the atmosphere: t (yr) of the various nuclides in a log-log coordinate system as shown in fig. 1.2.5., the values follow a straight line, i.e., a power curve:

$$T = 11.6 \cdot t^{1.7}. \qquad \text{(Eq.1.2.5.)}$$

The correlation between observed and calculated values ($r = 0.9979$) is highly significant.

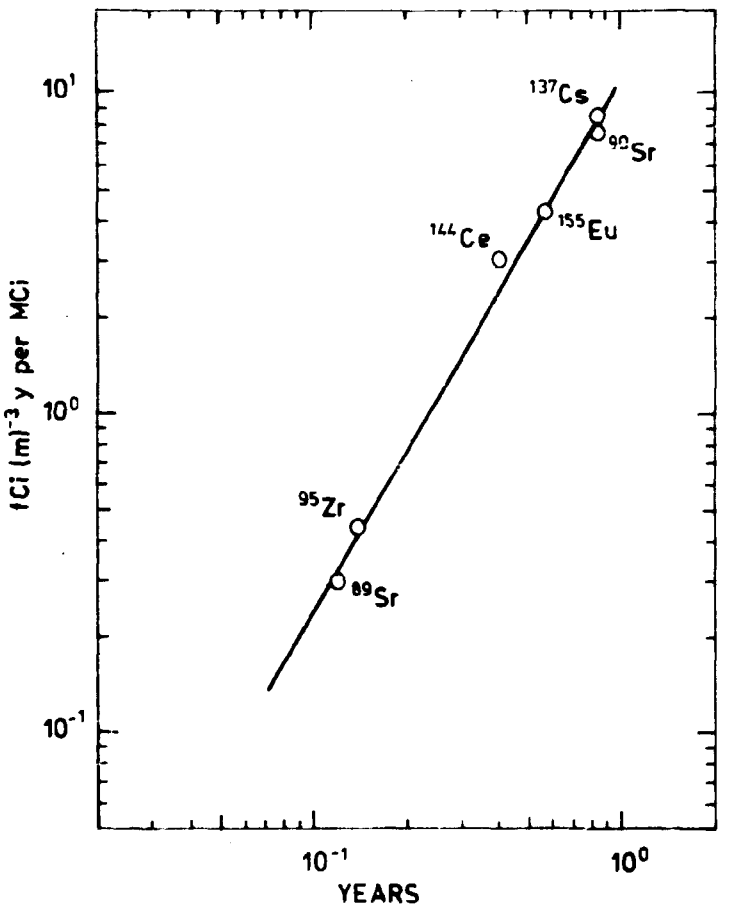

Fig. 1.2.5. The integral air concentrations (T) from 1 MCi injected into the northern hemisphere as a function of the effective half-lives (t) in the atmosphere: $T = 11.6 \cdot t^{1.7}$. T equals the transfer coefficient P_{01} from input to the atmosphere.

The above equation may be used for the calculation of the infinite time integrals of fallout radionuclides with effective atmospheric half-lives within the range of those included in the equation, i.e., between 0.1 and 0.9 years. Ruthenium 106 is such a radionuclide; the infinite time-integrated level was

in this case estimated at 1 pCi ^{106}Ru m^{-3} y. UNSCEAR (Un77) assumes a value of 0.50 for the ratio of the time integrals of the activity concentrations in air of ^{106}Ru and ^{144}Ce, hence the expected value was 0.5 · 2.3 = 1.15 pCi ^{106}Ru m^{-3} y, which agrees reasonably with the estimate.

Table 1.2.5. summarizes the time-integrated air concentrations for various fallout nuclides at Risø and the transfer factors from injections in the northern hemisphere. Short-lived radionuclides such as ^{131}I, ^{140}Ba, ^{141}Ce and ^{103}Ru do not become well mixed during periods comparable to the mean residence of an aerosol in the troposphere (Un77) and equation 1.2.5. does not apply in such cases. The transfer factors estimated from the equation for ^{103}Ru and ^{141}Ce were thus approx. twice as high as those actually found in the period 1971-1974.

Table 1.2.5. Estimates of time-integrated air concentrations, yields and transfer factors of various radionuclides in weapons debris collected in Denmark

Nuclide	Radioactive half-life	Effective half-life in the atmosphere in year	Nuclide/^{90}Sr at formation in thermonuclear tests	Integral air concentration fCi m^{-3} y until 1975	Total yield [c] MCi until 1975	Transfer factor fCi m^{-3} y per MCi	Observation period
^{54}Mn	312 d	0.43	-	149 [a]	54 [a,b]	2.8 [b]	1962-1966
^{89}Sr	50.5 d	0.12	185 (Un 77)	890	2960	0.3	1961-1963
^{90}Sr	28 y	0.83	1	121	16	7.6	1958-1975
^{95}Zr	64 d	0.14	225 (Harl 72)	1600	3600	0.44	1971-1974
^{103}Ru	39 d	0.095	380 (Harl 72)	700	6080	0.11	1971-1974
^{106}Ru	1 y	0.46	19.3 (Harl 72)	1000	309	3.1 [b]	
^{125}Sb	2.77 y	0.66	0.85 (Harl 72)	78	13.6	5.7 [b]	
^{131}I	8.04 d	0.021	1040 (Harl 72)		16640		
^{137}Cs	30 y	0.83	1.6 (Un 77)	222	25.6	8.7	1962-1975
^{140}Ba	12.8 d	0.034	1170 (Harl 72)		18720		
^{141}Ce	32.5 d	0.081	400 (Harl 72)	440	6400	0.069	1971-1974
^{144}Ce	285 d	0.41	47.6 (Harl 72)	2300	762	3.0	1961-1966
^{155}Eu	1.7 y	0.57	0.66 (III)	26 [a]	6 [a]	4.4	1961-1966
^{239} ^{240}Pu	2.44·10^4 y [e]	0.86	0.020 (Un 77)	2.87	0.32	9.0	

[a] 1962-1966.
[b] calculated values from Eq. 1.2.5.
[c] available as global fallout.
[e] ^{239}Pu.

During the 1961-1962 test series an estimated 630 Ci (Da66) *) of ^{32}Si was created. The transfer factor of a long-lived fallout nuclide was 9 fCi m^{-3} y per MCi, hence the expected

*) The ^{32}Si yield was previously estimated at 700Ci, (Da66), however, this estimate was based on an overestimated ^{90}Sr yield (cf.D.1.2.)

time-integrated air concentration was 5.7 aCi $^{32}Si\ m^{-3}\ y$. Measurements performed on May-June samples collected in 1962-1965 corresponded to 4.7 aCi $^{32}Si\ m^{-3}\ y$ (the annual concentrations were estimated from fig. 1.2.1.); considering that some bomb-produced ^{32}Si was still left in the atmosphere by June 1965, the two estimates are compatible.

1.3. Precipitation

Og Vinden kyssede Træet, og Duggen gravd Taarer over det, men det forstod Grantræet ikke.

GRANTRÆET

And the wind kissed the tree, and the dew wept tears over it, but this meant nothing to the fir tree.

THE FIR TREE

Normally, precipitation plays a predominant role in the transfer of radioactive debris from the atmosphere to the surface of the earth. Showers of rain can, however, both contaminate and decontaminate, by washing deposited debris off the surface of vegetation. In Greenland and the Faroes precipitation is often collected directly for drinking. This is usually not the case in Denmark with the exception of a few small islands.

Systematic measurements of ^{90}Sr were initiated in 1954 at Milford Haven in the U.K. (Cr60) and at the Health and Safety Laboratory (HASL) in New York (Ha58-78). Country-wide Danish studies at 11 locations began in 1962 (RRD62), and in the same year regular measurements were started of radiostrontium in precipitation samples collected in the Faroes at 2 stations (RRF62) and in Greenland at 5 stations (RRG62).

1.3.1. Variation with time and location

When radionuclide measurements are made of precipitation, the activity concentrations in the rain ($pCi\ l^{-1}$) as well as in the deposited activity ($mCi\ km^{-2}$) are usually determined; the fallout rate may be given as $mCi\ km^{-2}\ y^{-1}$.

The variability among years (table B.1.3.4.) of ^{90}Sr in Danish precipitation during 1962-1974 was 1.73 (fallout rate) and 1.86 (concentration). This is not significantly different

from the variabilities of the corresponding air concentrations (table B.1.2.3.). Among the months the variability in rain also corresponded to that observed in air ($CV_{p\ month} \sim 0.6$). In Faroese and Greenlandic precipitation the variability among years was identical to that in Danish rain. Among months, however, the Faroese rain showed a lesser variability than the Danish, i.e. the spring peak was less pronounced in the Faroes.

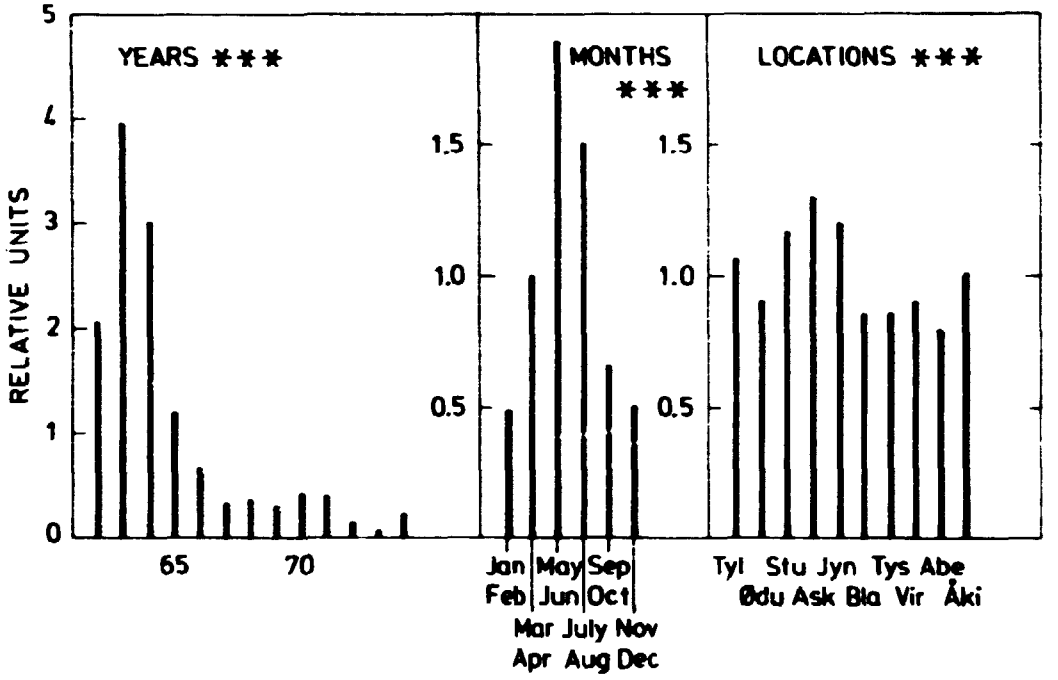

Fig. 1.3.1. The variation of ^{90}Sr Fallout rate with year, month and location assessed from precipitation samples collected at the Danish state experimental farms (fig. A.1.1.3.1.) 1962-1974. The bars indicate the Fallout rates relative to the grand mean: 0.55 mCi km^{-2}(2 months)$^{-1}$(= 1 at the relative scales).

The ^{90}Sr levels in rain decreased from the spring peak (in May-June) to the winter minimum (in November-December) with a half-life of approx. 60 days. The slope fitted the seasonal trend for ^{90}Sr in air from June to December. (fig. 1.2.1.).

The annual decay during the periods 1963-1967 and 1971-1973 corresponded to effective half-lives of 11 and 8 months, respectively. This is compatible with the 10-month half-life found for the annual ^{90}Sr and ^{137}Cs mean levels in air.

An anova (table B.1.3.3.) of the annual ^{90}Sr deposits in Denmark, the Faroes and Greenland showed no interaction between years and locations. The yearly ^{90}Sr deposition in mCi km^{-2} in the Faroes and Greenland may thus be estimated from the Danish data by multiplying by the mean ratios between the Faroese (or Greenlandic) and the Danish fallout levels. The following ratios were determined from data collected during the period 1962-1974:

Faroes (mean of Thorshavn and Klaksvik)/Denmark = 2.08
West Greenland (Godthåb)/Denmark = 0.75
East Greenland (Kap Tobin)/Denmark = 0.47

These ratios are used throughout the text where precipitation data are missing.

Within years, Danish data should not be used for the estimation of Faroese and Greenlandic fallout, as anovas showed significant interactions between months and locations.

The local variation among the Danish state experimental farms was highly significant for fallout rate (table B.1.3.2.), but in general not significant for pCi $^{90}Sr\ l^{-1}$ (Table B.1.3.1.). The local variability of fallout rate was 0.16, which corresponded to that of mm precipitation among the farms. We may expect the ^{90}Sr deposition to be proportional to the amount of precipitation in meters (Lu71, Me59-77):

$$\sum_{1962}^{1974} \text{mCi } ^{90}Sr\ km^{-2} = 5.3 + 4.9 \sum_{1962}^{1974} \text{m} \qquad \text{Eq. 1.3.1.}$$

As the regression line intercepts the ordinate axis at 5.3 mCi $^{90}Sr\ km^{-2}$, this figure is an estimate of the fallout for zero precipitation. This amount of so-called "dry fallout" corresponded to 11% of the total mean fallout in the period. The actual amount of dry fallout was not necessarily 11% because the rain collectors may have shown an efficiency with respect to collecting dry fallout that differed from that of the surface of the earth (Me56, Cham60). Studies of accumulated

^{90}Sr in Danish soil have, however, shown (cf.1.6.1.) that the amounts of ^{90}Sr found in the soil throughout the years have corresponded to the levels to be expected from the precipitation collectors. It is therefore concluded that, with respect to the estimation of total ^{90}Sr fallout, the rain-bottles seem to have been adequate. In 1957-1960 HARDY et al. (Ha62) found that, for an annual mean precipitation corresponding to that in Denmark, approx. 1/3 of the total ^{90}Sr fallout up until 1960 had been received as dry deposit. HARDY concluded that the specific activity of the precipitation was independent of the amounts of precipitation when the contribution from dry deposit was taken into account. In Denmark the contribution from dry deposit was too low to produce a significant local variation of the ^{90}Sr concentrations in rain water within the various years of observation.

Using Eq.1.3.1. for an estimate of the ^{90}Sr deposition in the Faroes and Greenland (Du75, Da63-76), the mean ratio between observed and estimated values became 0.73 ± 0.11 (1 SD), which is significantly less than unity. As air concentrations decrease with increasing latitude (Ha58-78), this was also to be expected for the concentrations in precipitation; we may thus expect a lower ^{90}Sr deposition in Greenland and the Faroes than in Denmark for the same amount of precipitation.

Dry deposition is of special importance in connection with direct contamination of vegetation. In lichen (D.3.4.4.) it was observed that the concentrations of radionuclides were more or less independent of variations in precipitation, but primarily related to the rain-water concentrations. During rainfall nuclear debris from the atmosphere is carried down to the vegetation cover, but at the same time the rain washes off the deposit on the vegetation. The two effects may offset each other, so that dry deposit only determines the contamination of plants (Wi67).

If it, according to Eq.1.3.1., was assumed that the dry depositon from 1962-1974 was 5.3 mCi $^{90}Sr\ km^{-2}$, this corresponded to a mean rate of dry deposition of $1.3 \cdot 10^{-5}$ pCi $^{90}Sr\ m^{-2}\ sec.^{-1}$. The mean air activity was $6.4 \cdot 10^{-3}$ pCi $^{90}Sr\ m^{-3}$ during

1962-1974. Thus the deposition velocity (Cham60) of fallout ^{90}Sr during 1962-1974 became:

$$V_g \text{ (m sec}^{-1}) = \frac{1.3 \cdot 10^{-5}}{6.4 \cdot 10^{-3}} \text{ m sec}^{-1} = 2 \cdot 10^{-3} \text{ m sec}^{-1}.$$

SMALL (Sm60) found for gross beta fallout activity collected in vessels during 1956-1959 deposition velocities ranging from $(2\text{-}34) \cdot 10^{-3}$ m sec^{-1}. PEIRSON et al. (Pei65) found during 1960-1964 a deposition velocity for global fallout in the range $(1\text{-}3) \cdot 10^{-3}$ m sec^{-1}. In case of tropospheric fallout V_g was nearly an order of magnitude higher. In his studies of dry deposition of fallout ^{137}Cs on alfalfa, WILSON (Wi69) reported a deposition velocity of $6 \cdot 10^{-3}$ m sec^{-1}, but for radon daughters the median deposition velocity on alfalfa was only $1 \cdot 10^{-3}$ m sec^{-1}; this value was close to those reported by CHAMBERLAIN (Cham60) for submicron particles in wind-tunnel experiments. The present value of $2 \cdot 10^{-3}$ m sec^{-1} for fallout ^{90}Sr was compatible with the above mentioned findings.

CHAMBERLAIN (Cham60) found in experiments that deposition velocity increased rapidly with particle size in the micron diameter range. The results of Risø's studies of the variation of specific radiostrontium activity with altitude in the 120 m high meteorological tower (I, II) were compatible with this observation. It appeared that the specific activity of precipitation samples increased with sampling height. The phenomenon was ascribed to increasing amounts of dry deposition in the rain-bottles located at the top of the tower, which was partly a result of the higher wind speeds and consequently higher deposition velocity here than at the bottom. It was furthermore demonstrated that the $^{89}Sr/^{90}Sr$ ratio increased with altitude. This is intelligible if the ^{89}Sr in global nuclear debris was generally attached to larger particles than was ^{90}Sr, because the ^{89}Sr was deposited with relatively fresh debris that settled first, while ^{90}Sr also occurred in old debris on fine particles.

1.3.2. Relations between concentrations in air and precipitation

The observed washout or scavenging ratio, W_o, is the ratio

between the concentrations of a given nuclide in precipitation and in air. The units used for W_o differ in the literature (En71); we shall apply:

$$W_o = \frac{\text{pCi } l^{-1} \text{ water}}{\text{fCi } m^{-3} \text{ air}}. \qquad \text{Eq.1.3.2.}$$

During the period 1960-1972 corresponding samples of air and precipitation were collected monthly at Risø and analyzed for ^{90}Sr; ^{89}Sr was analyzed in 1961-1963 and ^{137}Cs in 1960-1961 and 1965-1966. The overall mean of the 285 monthly scavenging ratios was estimated by means of VAR-3 (cf.B.3.) at 0.99, and the total coefficient of variation (cf.B.4.) was 0.51. The anova showed that the variation among nuclides was not significant. The variations among months as well as among years were, however, significant. The seasonal variation thus showed a maximum in January-February and a minimum in August-October. Several reasons can be suggested for such a difference. Firstly, during winter the relative amount of dry fallout, which was included in the precipitation samples, was higher than during the summer, due to the lower amounts of precipitation. Secondly, snow may be more efficient than rain as a scavenger of activity from the air. Thirdly, the absolute humidity of the air was lowest during the winter and, according to ENGELMANN (En71), the washout ratio should vary inversely with the absolute humidity. The annual variation of the washout ratios showed minima at the beginning and at the end of the period. There was no obvious explanation for this observation.

A comparison with the UK data on ^{137}Cs collected since 1954 at Chilton (Ae58-74) showed that the Danish annual mean washout ratio was approx. 1.5 times higher than the corresponding washout ratio observed in the UK. The difference between the two ratios was highly significant. Monthly scavenging ratios for ^{90}Sr determined from September 1969 to July 1970 in Arkansas varied between 0.55 and 1.45 with a mean value of 0.80 (No73). Washout ratios from six locations in the USA (En71) collected in 1962-1964 showed local mean values varying from 0.26 to 1.32. The Risø mean for the same period was 1.14. It thus seems that local variations in W_o are to be expected. There are,

however, no clear explanations of such variations, although absolute humidity and precipitation rate, as proposed by ENGELMANN (En71), seem to have some influence.

The total deposition of ^{90}Sr in Denmark was estimated from the precipitation data obtained at the state experimental farms. As the data were incomplete before 1962, the HASL data for New York (Ha58-78) were applied considering that the mean ratio between ^{90}Sr fallout in Denmark and New York was 0.7 in the period 1962-1974 (D.1.3.2.). The total amount of ^{90}Sr deposited since nuclear tests were started and up until 1974 was 73 mCi km^{-2} for Denmark (81.3 for Jutland and 64.8 for the Islands). These estimates may be compared with the UNSCEAR estimate of 79 mCi ^{90}Sr km^{-2} for the 50-60°N latitude band (Un77). We may, however, also calculate the total ^{90}Sr deposited in Denmark from the Danish air data (1.2.1.). According to these data the integral air level was 121 fCi ^{90}Sr m^{-3} y; as the observed washout ratio was 0.99, the time-integrated rain concentration became 120 pCi ^{90}Sr l^{-1} y. The mean precipitation at the state experimental farms was 628 mm y^{-1} (D.1.3.3.), thus the total deposition was 75 mCi ^{90}Sr km^{-2}, which is close to the above estimate.

1.4. Fresh water

Da hun igjen var klædt og havde flettet sit lange Haar, gik hun til de sprudlende Væld, drak af sin hule Haand og vandrede længere ind i Skoven, uden at vide hvorhen.
DE VILDE SVANER

When she was dressed again and had plaited her long hair, she went to the bubbling spring, drank from her cupped hands, and then wandered further on into the wood without really knowing where she was going.
THE WILD SWANS

Fresh-water systems are either lenitic, e.g. lakes and reservoirs, or lotic, e.g. streams and ground water (ground water may be considered as a subterranean stream). Radioactive contamination of fresh waters originates partly directly from the atmosphere, mostly via precipitation, partly indirectly from the soil, i.e., from older deposits that have been washed out. Nuclear installations may release radionuclides directly to fresh-water recipients. The transfer of radiocontaminants from fresh water to humans may either be directly through drinking water, or indirectly through animals and vegetation whose habitat is fresh water. Finally, fresh water is used for

irrigation and for drinking water for animals and may as such contaminate terrestrial food chains.

Systematic surveys of fresh water samples have been concentrated on ^{90}Sr assays of drinking water. HASL (Ha58-78) started such a study in 1954 on New York City tap water. The ^{90}Sr levels in a countrywide collection of samples of Danish ground water have been followed since 1961 by Risø (RRD61). Drinking water samples have been collected regularly in the Faroes and Greenland since 1962 and analyzed for ^{90}Sr (RRF62, RRG62).

The ^{137}Cs levels in fresh water are generally lower than the ^{90}Sr concentrations, because ^{137}Cs is retained to a larger degree than ^{90}Sr in soil minerals. (cf.1.6.)

1.4.1. Danish ground water

Radionuclides in ground water may reach man with the drinking water, which in Denmark is generally ground water. Irrigation of crops may be another way of exposure. Ground water passes through the soil layers, which act as a filter and a kind of ion exchange column. Clay minerals, which are abundant in Danish soils, are particularly efficient ion exchange media for several radionuclides contained in the water seeping through the soil. Eight of the nine borings used since 1961 for ground-water samples have in fact contained very low ^{90}Sr concentrations. The anova showed a highly significant variation among years (fig.1.4.1.1.). Although the maximum coincided with the maximum fallout rate in 1963, the variability among years of the ^{90}Sr concentrations in ground water (table B.1.4.1.) was significantly lower ($CV_{p\ year} = 0.57$) than the variability in rain water ($CV_{p\ year} = 1.8$). This suggested that although some ^{90}Sr may be rapidly transferred to the ground water, the concentrations also depend on the accumulated ^{90}Sr in the soil, which during the period of observation displayed a lower variability among years than the fallout rate (cf.1.6.).

The variation among the 8 localities was also highly significant (fig.1.4.1.1.). The local variability was 0.49, i.e. significantly higher than that observed for precipitation, because the

ground-water activity depended upon the ability of the surrounding soil layers to retain the radionuclides contained in the percolating water, rather than on local variations in the rain-water concentrations.

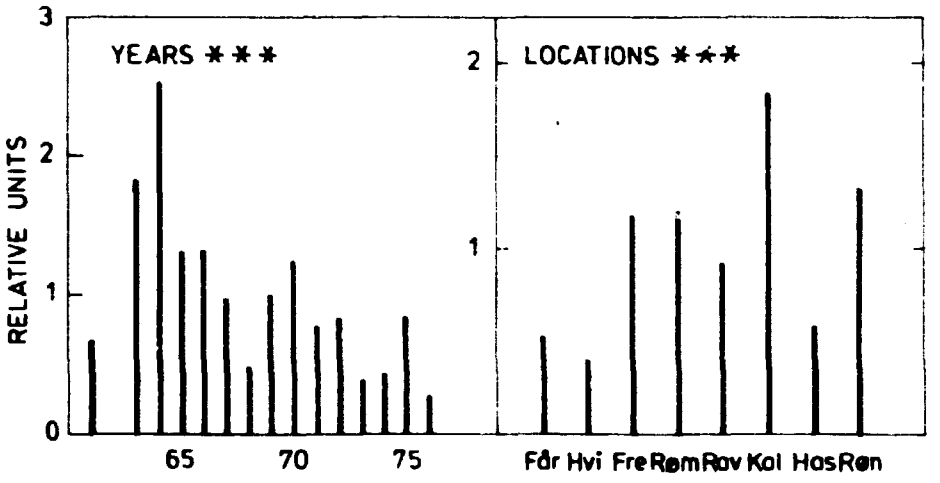

Fig. 1.4.1.1. The anual and local variation of ^{90}Sr in ground water collected in 1961-1976 from 8 borings in Denmark (cf. fig. A.1.1.4.1.). The bars indicate the concentrations relative to the grand mean: 14.7 fCi l^{-1} (= 1 at the relative scale).

The ^{90}Sr concentrations in Danish ground water may be related to the fallout rate and the accumulated fallout. The prediction models (table C.1.4.1.) show that the radioecological sensitivity of ground water to ^{90}Sr contamination from fallout was, as expected, extremely low: $7.4 \cdot 10^{-3}$ pCi ^{90}Sr l^{-1} y per mCi ^{90}Sr km^{-2}. The infinite time integral of ^{90}Sr in Danish ground water from nuclear weapons testing became $73 \cdot 7.4 \cdot 10^{-3}$ = 0.54 pCi ^{90}Sr l^{-1} y (cf. also D.1.4.1.).

Right from the start of the ground-water sampling programme one of the nine locations, Feldbak, has steadily shown relatively high ^{90}Sr levels. Furthermore, the levels have increased throughout the years, while the other eight locations have on the whole shown decreasing concentrations since 1963. Figure 1.4.1.2. shows that the levels until 1974 followed an exponential increase. Assuming the maximum concentration in the Feldbak ground water was reached in 1974, and from then that it follows the radioactive decay of ^{90}Sr, the maximum infinite time integral became 73 pCi ^{90}Sr l^{-1} y. This arose from a total

deposit in Jutland of 81.3 mCi $^{90}Sr\ km^{-2}$. Hence the maximum radioecological sensitivity of Feldbak ground water became: 0.90 pCi $^{90}Sr\ l^{-1}$ y per mCi $^{90}Sr\ km^{-2}$, or 2 orders of magnitude higher than that of the other ground-water samples.

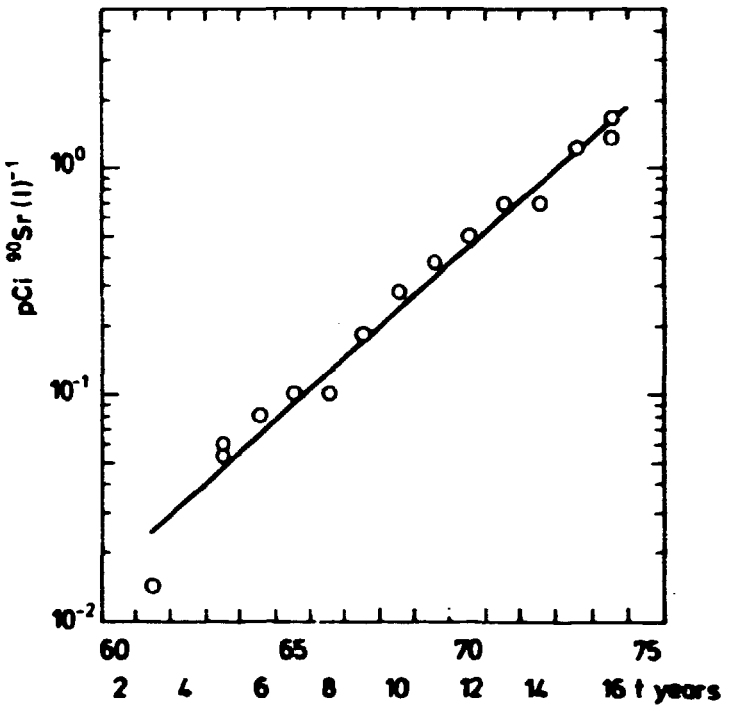

Fig. 1.4.1.2. Strontium-90 in ground water collected at Feldbak (cf. fig. A.1.1.4.1.) in 1961-1974. The concentrations increased exponentially:

$$pCi\ ^{90}Sr\ l^{-1} = 0.0093\ e^{0.32\ t}.$$

The infinite time integral (cf. the text) was:

$$\int_0^{17} 0.0093\ e^{0.32\ t} dt + \int_0^{\infty} 1.65\ e^{-\frac{\ln 2}{28}\ t} dt = 73\ pCi\ ^{90}Sr\ l^{-1} \cdot y.$$

The higher levels and the deviating time pattern found at Feldbak resulted from very poor filtering strata at this location. The filtering material consisted of sand which, compared to clay, only has a rather low cation exchange capacity. The ^{90}Sr layer deposited from precipitation therefore moved relatively rapidly through the filtering layers at Feldbak, and this explains the increasing levels.

1.4.2. Danish streams

The radionuclide content in stream water originates from direct deposition in streams, from activity contained in springs (ground water) and from the washing out of activity retained by soils and crops. In Denmark streams may be used for irrigation, and they are the habitat of fresh-water fish such as trout, but the consumption of fresh-water fish by the general Danish population is very modest.

An anova of the ^{90}Sr levels in stream water did not demonstrate any significant time variation during the period of systematic sampling from 1971-1975. However, 3 stream samples collected in Zealand in 1964 showed a mean of 2.0 $\pm$ 0.6 pCi $^{90}Sr\ l^{-1}$, as compared with 0.4 pCi $^{90}Sr\ l^{-1}$ from the same area of the country in 1970-1973.

The ^{90}Sr concentrations in stream water were thus undoubtedly related to the fallout rate. The anova also showed a significant local variation (fig.1.4.2.). The local variability was 0.43, i.e., nearly the same as found for ground water; it was thus again soil characteristics and other local environmental factors rather than the possible variations in rain-water concentrations that determined the local variability.

In December 1970 we undertook a study of the possible variation between the ^{90}Sr levels near stream sources and outfalls (RRD70). Six streams were measured and no systematic difference between the two positions was observed. Even the two largest streams in the study (Gudenå and Susså) showed no significant difference between the source and the outfall. As the ^{90}Sr concentrations of the stream water were 10-100 times higher than the ground water levels, it was evident that the main part of the ^{90}Sr in the streams came from run-off from the uppermost soil layers and from direct precipitation. In Denmark approx. 12 $km^3\ y^{-1}$ are run-off with lotic waters to the sea, corresponding to approx.45% of the annual precipitation (the remainder evaporates) (Je69). If the mean ^{90}Sr content in stream water for 1971-1975 (0.35 pCi $^{90}Sr\ l^{-1}$) was assumed representative of the ^{90}Sr concentration in run-off from the entire country, the mean annual amount of ^{90}Sr in the run-off during 1971-1975 is estimated at: $0.35 \cdot 10^{-12} \cdot 12 \cdot 10^{12} = 4.2$ Ci. As the accumu-

lated ^{90}Sr in Danish soil in 1971-1975 (RRD59-76) amounted to $55 \cdot 10^{-3} \cdot 43{,}069 = 2369$ Ci, the annual removal of ^{90}Sr becomes 1.8% of the accumulated fallout. During the periods with high fallout rates, when a substantial part of the fallout was present in the upper few centimetres of the soil, the run-off may have been somewhat higher than when the activity had been displaced down to a median depth of, say, 10 cm - a situation close to that in 1971-1975. (cf.Fig.1.6.2.)

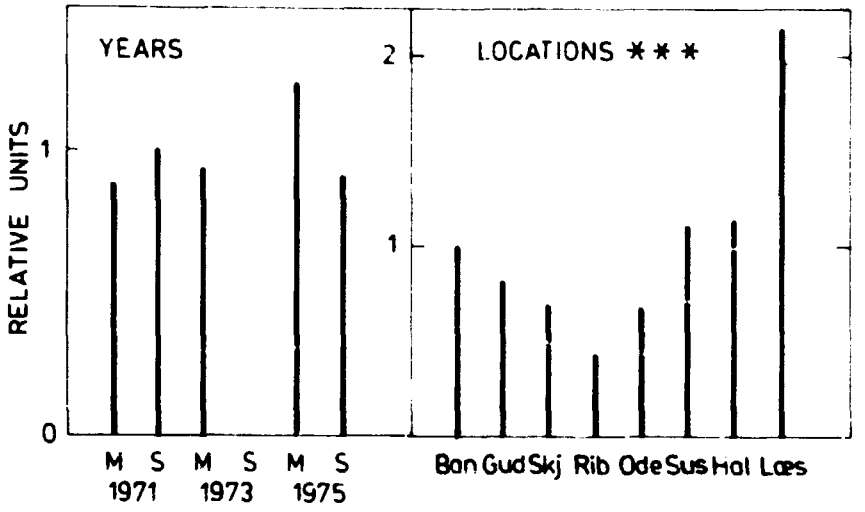

Fig. 1.4.2. The annual and local variation of ^{90}Sr in Danish stream water collected 1971-1975 (cf. fig. A.1.1.4.2.). The bars show the concentrations relative to the grand mean: 0.347 pCi ^{90}Sr l^{-1} (= 1 at the relative scales).

As in the case of ground water, prediction models were calculated for ^{90}Sr in Danish stream water (table C.1.4.1.). The radioecological sensitivity was estimated at 0.33 pCi ^{90}Sr l^{-1} y per mCi ^{90}Sr km^{-2}, and the infinite time integral from the ^{90}Sr deposition by 1974 (73 mCi ^{90}Sr km^{-2}) became 24.4 pCi ^{90}Sr l^{-1} y. The rate term in the prediction model (0.17 d_i) represented the contribution from relatively newly deposited fallout, we denote it the instant run-off The last terms (0.0028 $A_{i-1\ (6)}$ and 0.0035 $A_{i-1\ (28)}$) represented the so-called delayed run-off i.e. old debris eroded from the soil in the drainage area of the stream. From the total ^{90}Sr deposit of 73 mCi km^{-2}, the infinite time integral of instant run-off became 12.4 pCi ^{90}Sr l^{-1} y and the delayed, 12 . For the period 1950-1974, the total run-off in Denmark was estimated at 4.6 mCi ^{90}Sr km^{-2} (D.1.4.2.), corresponding to an annual average

removal of approx.5%, which is higher than the above estimate of 1.8%, but, as mentioned above, an annual run-off of 1.8% was an underestimate for freshly deposited ^{90}Sr. In a study of US streams made in 1959-1966, MENZEL (Me74) found an annual erosion of accumulated ^{90}Sr varying between 1.7 and 7.5%, and YAMAGATA et al. found 3.1% for ^{90}Sr and 0.6% for ^{137}Cs (Ya63) in 1962 in Japan.

1.4.3. Danish lakes

The total area comprised by lakes in Denmark is approx.550 km^2 (La69), or 1.3% of the total area. Lake water is to a limited extent used for drinking water, e.g., in the Copenhagen region. The lakes are the habitat of fresh-water fish but, as mentioned above, there is little consumption of these fish in Denmark.

The lake water was collected together with the stream water in 1971, 1973 and 1975. The anova showed no significant variation with time of the ^{90}Sr concentrations. A sample collected in June 1963 from Arresø contained 4.8 pCi $^{90}Sr\ l^{-1}$, or 3.6 times more than the corresponding mean level in 1971-1975. As the ^{90}Sr concentrations in rainwater decreased by a factor of approx.20 from 1963 to 1971-1975, this suggests that the activity in the lake water not only depended upon the fallout rate but also on the accumulated ^{90}Sr.

The local variation among the ^{90}Sr concentrations in lakes was highly significant. The variability was 0.83, i.e., higher than that observed for lotic waters. The explanation lies in the fact that some lakes have little exchange of water (e.g. Nors Sø), while others (e.g. Flyndersø) may be nearly lotic because streams flow through them. Such lakes may in the present context be regarded as a "widened stream", and the ^{90}Sr concentrations in such "lotic" lakes may be as low as in stream water, whereas the lakes with little exchange of water display relatively high ^{90}Sr concentrations (fig.1.4.3.).

The prediction model (table C.1.4.1. No 6) was based on relatively few observations, but the correlation coefficient between observed and calculated values was highly significant. The radioecological sensitivity was 1.1 pCi $^{90}Sr\ l^{-1}$ y per

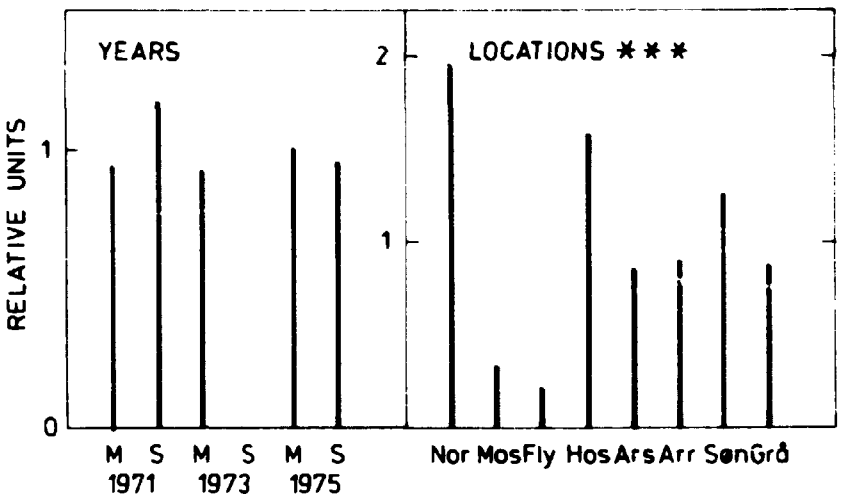

Fig. 1.4.3. The annual and local variations of ^{90}Sr in Danish lake water (cf. fig. A.1.1.4.2.) collected in 1971-1975 in March and September. The bars show the concentrations relative to the grand mean 1.35 pCi l^{-1} (= 1 at the relative scales).

mCi ^{90}Sr km^{-2}, i.e., 3 times higher than for streams. Using a similar prediction model, CARLSSON (Ca76) found a transfer factor for ^{137}Cs of 0.73 pCi ^{137}Cs l^{-1} y per mCi ^{137}Cs km^{-2} for a Swedish dysoligotrophic lake. The agreement between the two figures may be fortuitous as ^{90}Sr and ^{137}Cs do not behave in the same way in freshwater systems (cf.1 6.3.), and because the Danish lakes were not dysoligotrophic but rather eutrophic (La69).

1.4.4. Danish drinking water (tap water)

The main source of tap water in Denmark is ground water. Surface water, however, is used in increasing amounts, especially in the Copenhagen region.

An anova of the ^{90}Sr data on tap water collected from the whole country since 1965 showed a significant time variation. The levels in 1965 and 1971 were thus higher than the levels in 1970 and 1973. However, in contrast to ground water, it was not possible to observe a general decreasing trend in the levels.

The variation between locations was highly significant. Bornholm, East Jutland and Lolland-Falster showed 4-5 times higher levels than the rest of the country. The local variability was 0.79 (table B.1.4.1.); the high variability was due to the contribution of surface water to the potable water at certain locations. Tap water from Copenhagen, which was not included in the anova, contained significantly more ^{90}Sr than the drinking water from the rest of the country.

Reliable prediction models for tap water were difficult to set up because the data from year to year showed great variations that seemed uncorrelated to fallout data as well as to accumulated fallout. An inexplicably high value from 1971 was especially annoying in this respect. The prediction models (table C.1.4.1.) disregarded this value. The radioecological sensitivity became $4.9 \cdot 10^{-3}$ pCi ^{90}Sr l^{-1} y per mCi ^{90}Sr km^{-2}, which was of the same order as that estimated for ground water. This suggested that borings, such as that in Feldbak which showed relatively high ^{90}Sr levels, did not play any important role for the supply of Danish tap water.

If the Copenhagen tap water was included, the population weighted mean sensitivity of Danish drinking water became $9 \cdot 10^{-3}$ pCi ^{90}Sr l^{-1} y per mCi ^{90}Sr km^{-2}, because the mean ^{90}Sr concentration in drinking water from Copenhagen was 4 times that in the remaining part of the country. As the annual per caput drinking water intake by the Danish population is 548 l, the infinite time-integrated ^{90}Sr intake from drinking water became $9 \cdot 10^{-3} \cdot 548 \cdot 73 = 360$ pCi ^{90}Sr, which was less than 2% of the total ^{90}Sr intake of a Danish individual (cf.4.2.2.).

Although drinking water was thus unimportant as a source of ^{90}Sr, it was a main contributor to the stable Sr intake. The usual mean concentration was about 6 mg Sr (g Ca)$^{-1}$, or 0.5 mg Sr l^{-1}; drinking water from Lolland-Falster, however, contained 28 mg Sr (g Ca)$^{-1}$, or 2.8 mg Sr l^{-1}, and in Zealand the level was 17 mg Sr (g Ca)$^{-1}$, or 1.7 mg Sr l^{-1}. The occurrence of coelestin ($SrSO_4$) in the underground of the south-eastern part of the country (Ra75) may explain the higher stable Sr/Ca ratios found in Lolland-Falster; the inflow of sea-water may in some borings be another factor contributing to the higher

stable Sr to Ca ratios, because the mg Sr/g Ca ratio in sea water is normally higher than in fresh water (Od50). In a study of Ca and Sr in raw and tap water from 50 cities located throughout the United States, ALEXANDER et al. (A154) found that the highest Sr concentrations were associated with waters from regions having soils containing a large proportion of easily leached soft lime carbonate. The highest Sr/Ca ratio in the U.S. study was similar to that observed for Lolland-Falster, but the mean content of stable Sr in US tap water was one third only of that in Danish tap water (Lolland-Falster and Zealand excluded).

1.4.5. Faroese tap water

Faroese drinking water is obtained from surface water. An anova of the ^{90}Sr data collected in 1962-1974 showed a highly significant variation among years. The variability was 0.55, and thus significantly less than that observed in Faroese precipitation ($CV_{p\ year}$ = 1.78), suggesting that the ^{90}Sr concentration in Faroese tap water also depended upon the accumulated fallout.

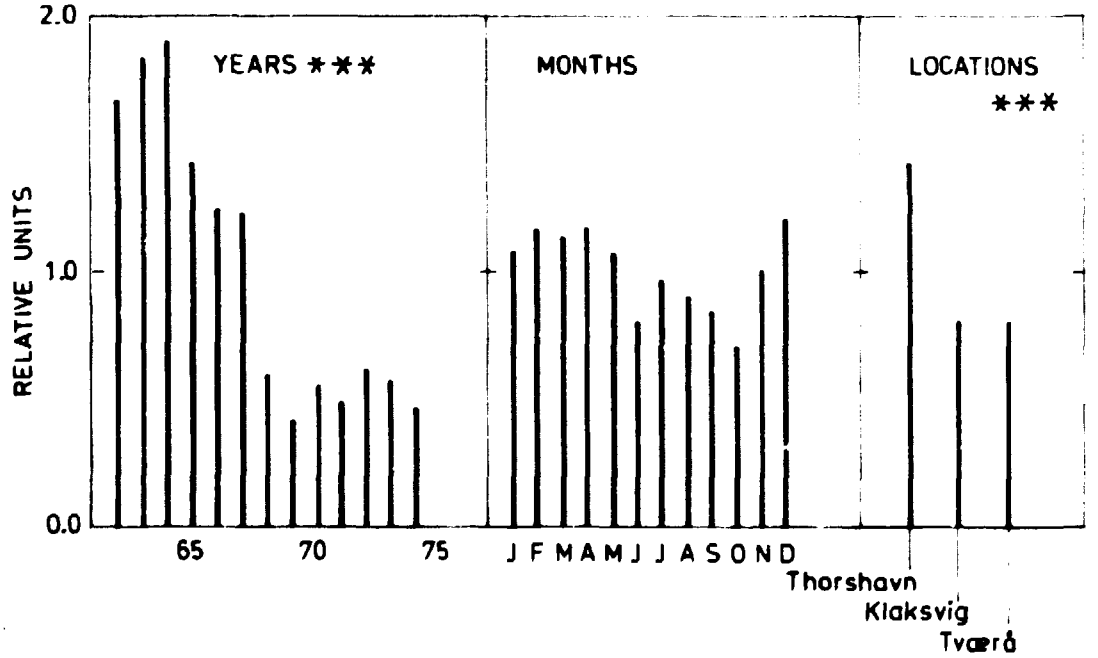

Fig. 1.4.5. The annual, monthly and local variation of ^{90}Sr in Faroese drinking water collected in 1962-1974 (cf. fig. A.1.1.3.2.). The bars show the concentrations relative to the grand mean 0.58 pCi l^{-1} (= 1 at the relative scale).

In 1962 (RRF62) and 1963 (RRF63) the $^{89}Sr/^{90}Sr$ ratios were determined in Faroese tap water. The measurements showed that the ratio in tap water was approx. half of that in precipitation from the same month. This suggested that approx. half of the ^{90}Sr in Faroese tap water had been obtained from recent precipitation.

The local variation was also highly significant (fig.1.4.5.). During the whole period the levels at Thorshavn were nearly twice as high as the ^{90}Sr concentrations at Klaksvig and Tværå, where the levels did not differ significantly from each other. It has been shown (RRF67) that the lower levels at Tværå, as compared with Thorshavn, coincided with a 2-3 times greater ion-exchange capacity for Sr of the Tværå soil.

Because of the pronounced local variations in the ^{90}Sr concentrations, prediction models were calculated for the water for each of the three locations (table C.1.4.1.). The radioecological sensitivity of the Thorshavn drinking water was nearly five times as high as that of the water from Klaksvik. The mean sensitivity of Faroese tap water was 0.11 pCi ^{90}Sr l^{-1} y per mCi ^{90}Sr km^{-2}. The infinite time-integrated level from a total deposition of 152 mCi ^{90}Sr km^{-2} became 17 pCi ^{90}Sr l^{-1} y, which was nearly 30 times higher than in Danish drinking water (0.66 pCi ^{90}Sr l^{-1} y). The infinite time integral in the Faroese tap water was similar to that observed for Danish streams (24 pCi ^{90}Sr l^{-1} y), but definitely lower than for Danish lakes (82 pCi ^{90}Sr l^{-1} y). The calcium concentration in Faroese drinking water was measured at 0.004 ± 0.002 g Ca l^{-1} in 1962 (RRF62), i.e. 5% of that in Danish drinking water.

1.4.6. Greenlandic drinking water

Drinking water in Greenland is obtained from two main sources: surface water from lakes, rivers or rain-water reservoirs, and meltwater from snow and (old) ice. The activity levels strongly depend upon the source of the water. The old ice thus has zero activity, while snow and surface waters may often have concentrations close to those found in precipitation.

An anova carried out on the yearly ^{90}Sr mean concentrations in Greenlandic drinking water from 1962-1974 showed a highly significant variation among years. The variability was 0.61 (table B.1.4.1.), or similar to that observed in Faroese drinking water but significantly lower than in Greenlandic precipitation (CV_p year = 1.75). As in the case of Faroese drinking water, we may conclude that the ^{90}Sr levels in Greenlandic drinking water did not solely depend on fresh fallout but also on older deposition of ^{90}Sr.

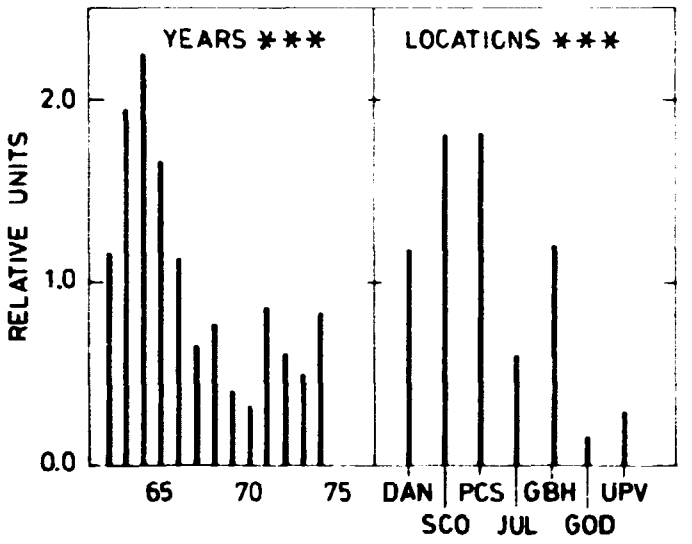

Fig. 1.4.6. The annual and local variation of ^{90}Sr in Greenlandic drinking water collected in 1962-1974 (cf. fig. A.1.1.3.3.).
DAN = Danmarkshavn, SCO = Scoresby Sund, PCS = Prins Christians Sund, JUL = Julianehåb, GHB = Godthåb, GOD = Godhavn, UPV = Upernavik. The bars indicate the concentrations relative to the grand mean 1.30 pCi l^{-1} (= 1 at the relative scale).

The local variability was 1.14 in Greenlandic drinking water (fig.1.4.6.), which value was higher than found in Danish or Faroese water. There were low levels at Godhavn and Upernavik, where snow and ice are used in the supply of drinking water, and the highest level was found in water from Prins Christians Sund where rain-water was collected in a reservoir and used directly for drinking. The local variation was thus a result of the extraction of the water used at the given location, rather than of local variations in waters of the same origin.

The prediction model for ^{90}Sr in Greenlandic drinking water (table C.1.4.1.) was based upon the samples received from the west coast of Greenland between Upernavik and Julianehåb. As the dat.. were incomplete, annual means calculated by VAR-3 (B.3.) were applied in the model. The fallout data used were those measured at Godthåb (cf.1.3.). The radioecological sensitivity was estimated at 0.52 pCi ^{90}Sr l^{-1} y per mCi ^{90}Sr km^{-2} and the infinite time exposure integral from a deposit of 55 mCi ^{90}Sr km^{-2} was 29 pCi ^{90}Sr l^{-1} y, i.e., higher than in Faroese and Danish drinking water. The lower temperatures in Greenland, which give permafrost at many locations, may prevent the soil from acting as an effective absorbing medium for ^{90}Sr and thus of removing the activity from Greenlandic fresh water.

1.5. Sea water

Langt ude i Havet er Vandet saa blaat, som bladene på den deiligste Kornblomst og saa klart som det reneste Glas.

DEN LILLE HAVFRUE

Far out at sea the water's as blue as the petals of the loveliest cornflower, and as clear as the purest glass.

THE LITTLE MERMAID

The anthropogenic radioactive contamination of the sea originates primarily from nuclear weapons testing. As approx.70% of the surface of the world consists of water, a substantial part of the global fallout has been deposited in the oceans. Furthermore, the sea is the ultimate recipient of run-off from the land masses. In local waters, contamination from peaceful uses of nuclear energy may predominate over fallout. This has, e.g., been the case in recent years in the North Sea, which is contaminated with radionuclides released from the reprocessing plants at Windscale and at Cap de la Hague.

Some radionuclides, e.g. ^{90}Sr, stay in solution in the sea, while others, e.g. plutonium and to some degree ^{137}Cs, settle in the sediments. The latter process is especially found in estuaries where the concentration of particulate matter is high and where lower salinities occur.

The marine environment is a habitat for animals and vegetation exploited by man, and studies of the radioactive contamination

of the sea were initiated already in 1946 (Se71) shortly after nuclear weapons tests began. Global contamination with ^{90}Sr has been studied in surface sea water from the Atlantic Ocean since 1954 by HASL and the Woods Hole Oceanographic Institution (WHOI) (Vol71). Risø has since 1962 measured the ^{90}Sr levels in sea water samples collected in inner Danish waters (RRD62) and in the waters along the coasts of the Faroes (RRF62) and Greenland (RRG62).

1.5.1. Inner Danish waters

In inner Danish waters a northward surface current carries the light Baltic water out through the Danish Sounds and Belts, and a southward bottom current transfers the heavy North Sea water to the Baltic Sea (He68). In 1963 the ^{90}Sr concentrations in the Baltic (Vo 1) and in the North Sea (Vol71) were approx. the same ($\sim$ 0.6 pCi ^{90}Sr l^{-1}), while the levels in inner Danish waters were somewhat higher ($\sim$ 0.9 pCi ^{90}Sr l^{-1}), because the rather intensive fallout rate at that time influenced the ^{90}Sr concentration of the shallow Danish waters to a larger degree than the deeper waters usually found in the North Sea and the Baltic Sea. Finnish data (Voi71) suggest that the situation persisted until about 1965 due to the relatively intensive fallout rate till then.

By 1966 the situation had changed. At that time the Baltic Sea contained the highest levels ($\sim$ 0.9 pCi ^{90}Sr l^{-1}) because run-off, fallout, and inflow from Danish waters in the preceding years had all contributed to an increase of the inventory of the Baltic. The water in the North Sea had in the same period been diluted to about 0.4 pCi ^{90}Sr l^{-1} by the low level ($\sim$ 0.2 pCi ^{90}Sr l^{-1}) Atlantic Ocean water. By 1966 the mean concentration of inner Danish waters had decreased to about 0.7 pCi ^{90}Sr l^{-1}.

In accordance with the above considerations, the lowest ^{90}Sr concentrations in 1963 would be expected in the low salinity samples, because the activity in these samples was diluted by the low activity water from the Baltic Sea. A similar dilution did not take place for the high salinity surface water from the Cattegat because the heavy North Sea water remains at the

bottom. Hence proportionality was found between salinity and ^{90}Sr concentrations in inner Danish waters in 1963.

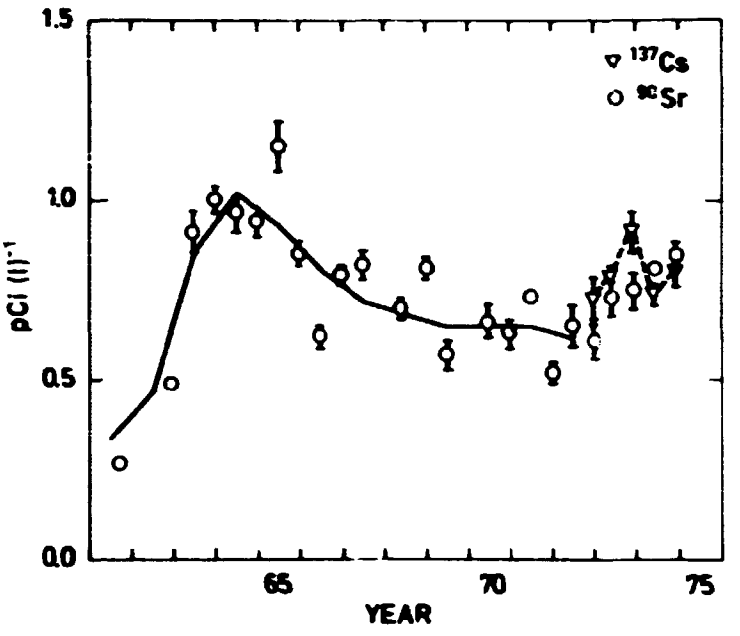

Fig. 1.5.1.1. Strontium-90 and ^{137}Cs in Danish surface sea water collected 1962-1974 in the inner Danish waters (cf. fig. A.1.1.5.). The values shown were the means of approx. 10 determinations (± 1 SE indicated). The full curve shows the predicted values (cf. Table C.1.5.1. No 1). The dotted curve connects the ^{137}Cs means.

After 1966 the low salinity Baltic water carried the highest ^{90}Sr concentrations, while the inner Danish waters - due to the lower fallout rates at that time and to the dilution with low activity North Sea water that had been increasingly mixed up in the water column - carried lower concentrations. An inverse relationship between salinity and ^{90}Sr concentrations in the inner Danish waters was therefore observed from 1966 (cf. table D.1.5.1.).

Equation no. 1 in table D.1.5.1. implied that the Baltic component (∿ 10%) of the inner Danish waters during 1967-1971 showed a mean level of 0.8 pCi $^{90}Sr\ l^{-1}$, while the North Sea component (∿ 34%) contained 0.3 pCi $^{90}Sr\ l^{-1}$. Since 1972 the equations have gradually changed. The ^{90}Sr concentration of North Sea water has nearly doubled since 1967-1971, while the ^{90}Sr concentration of Baltic water is essentially unchanged.

The equations for ^{137}Cs in table D.1.5.1. indicate that the activity levels in inner Danish waters are again approaching direct proportionality to the salinity. However, as compared to 1963-1965, the mechanism differs. In recent years the ^{90}Sr and ^{137}Cs levels in the North Sea have increased (fig.1.5.1.1.) due to the operation of reprocessing plants in the UK and in France (Je73, De71-77).

Inner Danish waters have thus shown an evident time variation of the ^{90}Sr levels, which were at first dominated by fallout but later influenced by releases from reprocessing plants indirectly to the North Sea. The local variations were, as a whole, obscured by interaction (fig.1.5.1.2.). For the period 1967-1972, the ^{90}Sr concentrations were inversely proportional to the salinity, and the levels found in the Baltic water were typically 2-3 times higher than that of the North Sea.

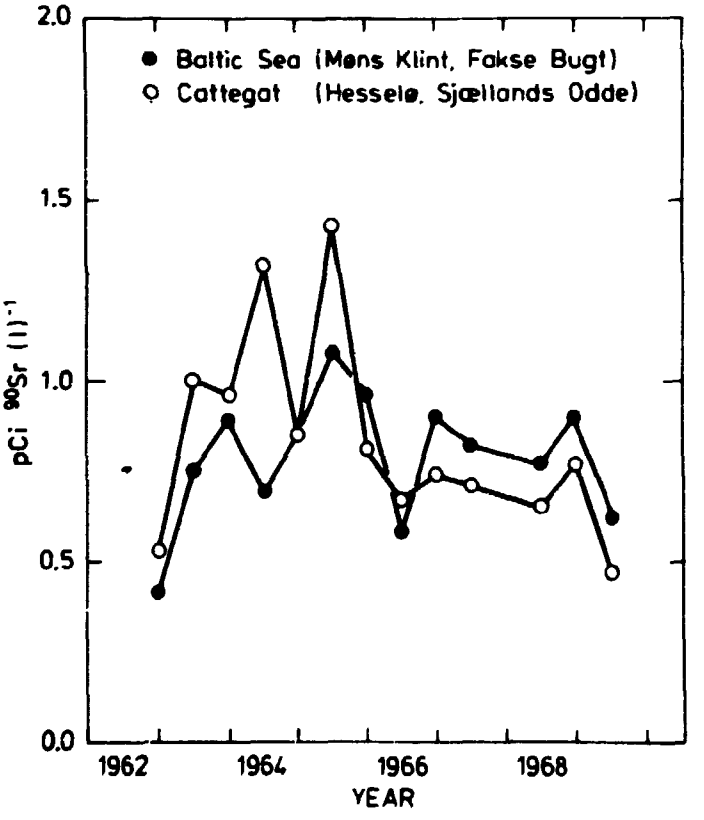

Fig. 1.5.1.2. Strontium-90 in Baltic sea water (salinity 10‰) and in Cattegat sea water (salinity 20‰) collected in 1962-1969 at two locations for each type of water (cf. fig. A.1.1.5.). Up until 1966 the "high salinity" stations in the Cattegat showed higher levels than the "low" salinity stations, but since then this situation has been reversed (cf. the text).

The variability among years (table B.1.5.1.) was 0.26 for ^{90}Sr in Baltic water (1961-1974) and 0.18 in North Sea water (1963-1974). The time variability of Danish sea water was low compared with the fallout rate ($CV_{p\ year}$ = 1.7), but of the same order as that observed for ^{90}Sr accumulated in the soil. Organisms acquiring their radionuclide contents from the sea usually display a low variability among years compared to most terrestrial organisms, because the radionuclide levels of the latter are more strongly influenced by the fallout rate.

During 1972-1974 the variability of ^{137}Cs among years in Baltic water was lower than that in North Sea water because the time variability of the ^{137}Cs concentrations in the North Sea was enhanced by the varying releases of ^{137}Cs from Windscale and Cap de la Hague.

Prediction models for ^{90}Sr concentration in inner Danish waters were calculated from the data on surface water collected in the period 1961-1972 (table C.1.5.1.). The radioecological sensitivity was estimated as the mean of the two models (nos. 1 and 2 in table C.1.5.1.), i.e., 0.35 pCi $^{90}Sr\ l^{-1}$. y per mCi $^{90}Sr\ km^{-2}$, and the infinite time-integrated ^{90}Sr level in Danish surface sea water (mean salinity 16‰) became 26 pCi $^{90}Sr\ l^{-1}$ y. As the mean Ca content in sea water with a mean salinity of 16‰ is 0.19 g l^{-1}, the radioecological sensitivity of Danish sea water may also be expressed as 1.9 pCi ^{90}Sr $(g\ Ca)^{-1}$ y per mCi $^{90}Sr\ km^{-2}$.

The mean ratio between ^{137}Cs and ^{90}Sr in surface water during 1972-1974 was 1.07. Assuming this ratio to be valid also for the period 1960-1972, which is compatible with Finnish data (Voi71), we may estimate the transfer coefficient for ^{137}Cs as $\frac{0.35 \cdot 1.07}{1.6}$ = 0.23 pCi $^{137}Cs\ l^{-1}$ y per mCi $^{137}Cs\ km^{-2}$ deposited. It is remarkable that the transfer coefficient for ^{137}Cs was approx. 2/3 of that for ^{90}Sr; the reason for this being the sedimentation of ^{137}Cs (cf.6.3.). The radioecological sensitivity to ^{90}Sr contamination of sea water was about one third of that of Danish lake water, which was partly due to the shorter residence time of ^{90}Sr in sea water than in most lakes.

1.5.2. North Atlantic ocean and Greenlandic waters

Since 1962 surface sea-water samples have been collected at Thorshavn and along the coasts of Greenland. These samples were supplemented by other samples taker by the M/S DANA on cruises to the Faroes and Greenland during 1962-1974. Anovas of the ^{90}Sr concentrations showed that the highest levels were found in East Greenland waters and the lowest in the Atlantic Ocean and the coastal Faroese waters. There was no significant difference between the Atlantic Ocean water and the Faroese water. With respect to ^{90}Sr content, we may thus consider the Faroese sea-water samples to be representative of North Atlantic Ocean water from latitude 60^{o} - $70^{o}N$ between the Faroe Islands and Greenland. The ^{90}Sr levels of West Greenland waters were not significantly different from those of North Greenland waters.

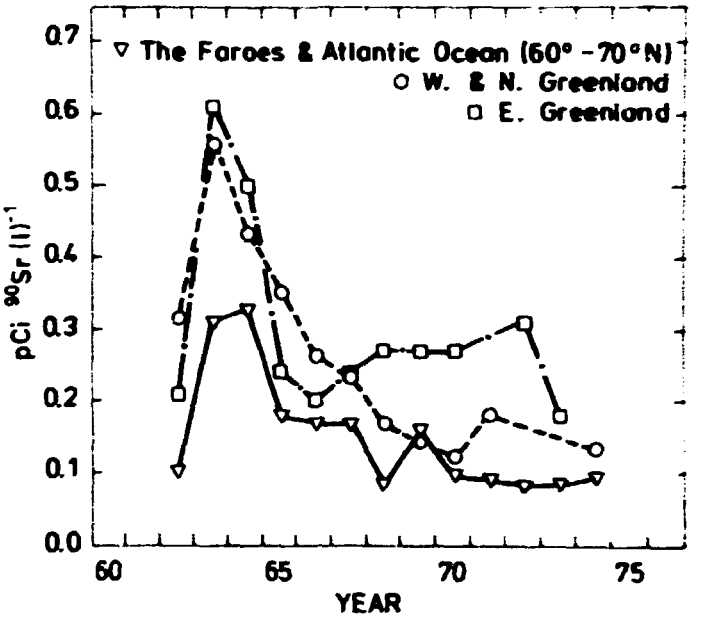

Fig. 1.5.2. Strontium-90 in North Atlantic surface sea water collected in 1962-1974 between and at the Faroes and Greenland.

Figure 1.5.2. depicts the interaction between time and locations. The East Greenland waters showed a diverging time variation; since 1967 the levels in East Greenland waters have thus been higher than in West and North Greenland waters. Sea-water samples from the GEOSECS (Vol73a) cruise to the Greenland Sea in August 1972 indicate that ^{90}Sr inventories are here greater

than expected. Samples from the East Greenland Polar current collected in 1966 (RRG66) by the DANA suggested that the ^{90}Sr levels in this current are a little higher than in the Irminger current. There seems no obvious explanation of the enhanced ^{90}Sr concentrations in the Polar current, nor of the higher levels generally found in samples from East Greenland waters (RRG62-76).

The variabilities among years (1962-1974) in North Atlantic and Greenlandic waters were 0.48 and 0.46, respectively, i.e., significantly higher than in Danish waters (table B.1.5.1.). This was a result of the higher dilution occurring in ocean waters than in shallow seas. In recent years some samples of Faroese (RRF62-76) waters have shown enhanced $^{137}Cs/^{90}Sr$ ratios (> 2), suggesting contributions of ^{137}Cs from Windscale to the North Atlantic Ocean (Ku78).

The radioecological sensitivity of North Atlantic surface water to ^{90}Sr contamination from fallout was low: 0.03 pCi ^{90}Sr l^{-1} y per mCi ^{90}Sr km^{-2} (table C.1.5.1.); the reason for this being the effective dilution by the large sea-water masses. Greenlandic coastal waters showed a sensitivity similar to that observed in inner Danish waters, i.e., an order of magnitude higher than in the North Atlantic Ocean water. The East Greenland waters were apparently more sensitive than the West Greenland waters, or they may have been subject to an unknown source of radioactive contamination.

The radioecological sensitivity of North Atlantic surface sea water to ^{137}Cs contamination was equal to that of ^{90}Sr, because the $^{137}Cs/^{90}Sr$ ratio in the ocean water was similar to that in nuclear debris.

DANA collected deep water samples on a number of occasions (cf. table D.1.5.2.). Concentrations of ^{90}Sr in the deep sea have been a most controversial subject (Vol71). BOWEN et al. (Bo60) were thus convinced that measurable ^{90}Sr (and ^{137}Cs) concentrations have been present below a depth of 1000 m in the North Atlantic since the beginning of the sixties, while BROECKER et al. (Bro66a) thought it safe to say that any finite amounts of ^{90}Sr and ^{137}Cs found prior to 1965 in samples from

below 1500 m were almost certainly due to undetected blanks. The present material showed that samples collected from about latitude 60-65°N, where the vertical mixing of the Atlantic Ocean is pronounced (He66), displayed ^{90}Sr concentrations that were definitely above background concentrations. A few samples collected from about latitude 40°N in 1966 showed lower levels, and for one location it was doubtful whether the levels below 2000 m were significantly above the detection limit.

From the DANA samplings in 1966, 1971 and 1973, the total integrated ^{90}Sr level in the water column from about 62°N in the North Atlantic (RRG73) was estimated at 146 ± 12 (1 SE) mCi ^{90}Sr km^{-2}, i.e. not incompatible with the accumulated ^{90}Sr fallout in the Faroes and in Prins Chr. Sund. During the GEOSECS cruise in 1972 (Vol73a) similar observations were made at other locations in the North Atlantic. But according to fallout studies carried out on 4 weather ships in the North Atlantic Ocean (Vol74, Vol73b), the expected fallout was only half of that actually found in the water column. The study of ^{90}Sr concentrations in deep water samples collected in the North Atlantic Ocean thus suggest an excess ^{90}Sr fallout in this part of the Ocean, which seems incompatible with the levels expected from fallout measurements on the weather ships. Various explanations of this phenomenon have been proposed. For example, it has been assumed that the amount of dry fallout was greater over the sea, or that sea spray activity scavenged the ^{90}Sr from the air. None of these mechanisms has so far been demonstrated in environmental (Bro66b) or in experimental studies (Vol74). Assuming a substantially greater fallout rate over the ocean than over land implies that the global inventory from weapons testing has been seriously underestimated (at least by a factor of two), or that the ^{90}Sr inventories on land have been overrated. According to VOLCHOCK (Vol74), both hypotheses seem unlikely. Measurements in the Pacific and Indian Oceans do not suggest these oceans to be depleted in ^{90}Sr as compared with the Atlantic Ocean.

1.6. Soil and sediment

Regnormen kom lige op af Højen, hvor den, Natten og Dage havde rodet i Jorden; den havde hørt en heel Deel, see kan den jo ikke, det elendige Dyr.
ELVERHØJ

"And I've had a talk with an earthworm I know," said a third lizard. "He was just coming up from the Hill, where for nights and days he had been rummaging in the ground, and he had overheard a great deal. Of course, he can't see, poor creature.
THE HILL OF THE ELVES

In the terrestrial environment soil is the final recipient of radioactive debris not removed by re-suspension or run-off to the sea. The ability of the soil to retain radioactive substances depends upon numerous environmental factors: soil characteristics, meteorological and hydrological conditions, man's use of the terrestrial environment, animal life and vegetation, and the physical and chemical properties of the radioactive debris. Crops may absorb radionuclides from the soil through their root systems; radioactive substances are then introduced into the food chains by so-called indirect contamination. The extent to which this happens also depends upon the factors mentioned above. Most of the deposited radioactive contamination, however, remains ion-exchanged or physically adsorbed in a soil rich in clay minerals.

The first systematic soil sampling for ^{90}Sr determination was carried out in 1955 in the United States (Has58). In the following years this soil programme was expanded by HASL to a global study of the accumulated ^{90}Sr levels in soil. Country-wide soil sampling aimed at estimating the accumulated ^{90}Sr in Danish soil was initiated by Risø in 1961 (RRD61).

1.6.1. Variation with time and location

In theory, the fallout A_n accumulated by the year n is the sum of the annual amounts of fallout d_i up till n corrected for radioactive decay:

$$A_n = \sum_{i=0}^{i=n} d_i e^{-\lambda_r (n-i+1)} \qquad \text{(Eq.1.6.1.)}$$

where λ_r is the decay factor (for ^{90}Sr: 0.025 yr^{-1}); i=0 in 1950, when fallout began. Equation 1.6.1. presumed that all fallout deposited in a year (i) had been received by the start

of the year. Although more than half of the long-lived global fallout is usually deposited in the first part of the year, the method of calculation applied will underestimate the accumulated fallout. For long-lived radionuclides, such as ^{90}Sr and ^{137}Cs, this underestimate is, however, immaterial.

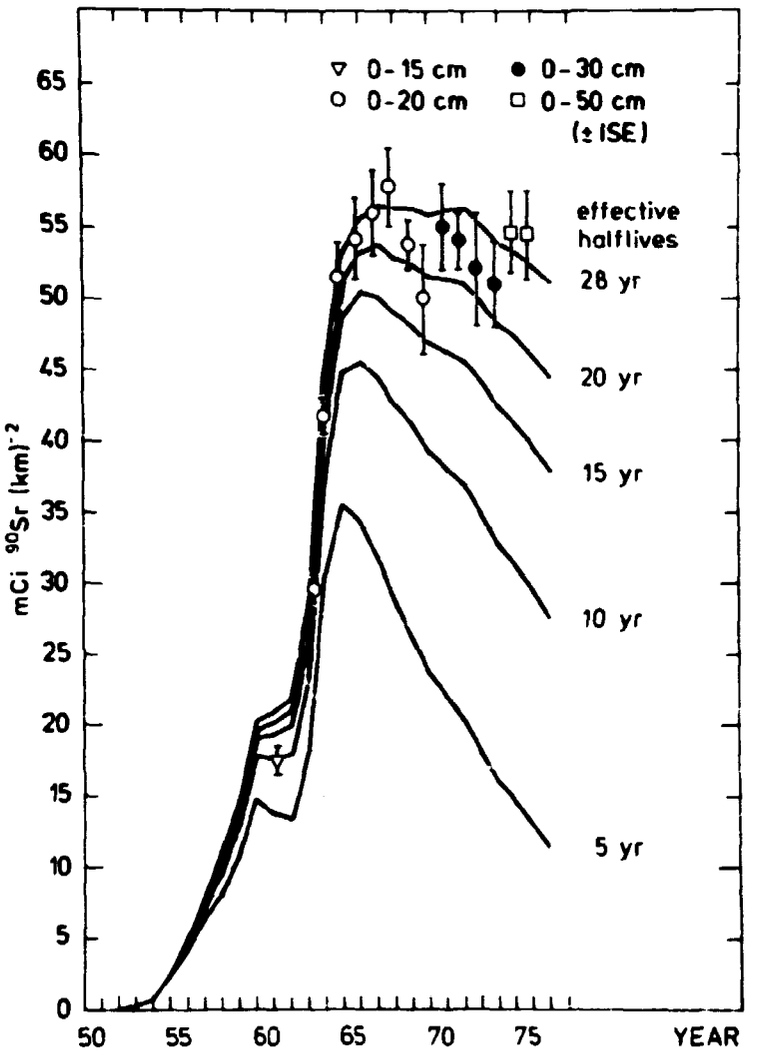

Fig. 1.6.1. The ^{90}Sr levels observed in Danish uncultivated soils collected in 1961-1975 at the state experimental farms (fig. A.1.1.3.1.) compared with the calculated levels (the curves) assuming various effective half-lives (cf. Eq. 1.6.1.) of the ^{90}Sr deposited. The correlation coefficients between observed and calculated levels were: 0.9825 for 28 yr, 0.9700 for 20 yr, 0.9289 for 15 yr, 0.7834 for 10 yr and 0.3587 for 5 yr.

Figure 1.6.1. shows a comparison between the levels observed in uncultivated soil and the calculated values. The upper curve was calculated from (Eq.1.6.1.) using the Danish precipitation data (1.3.) as d_i values. The underlying curves were analogous, assuming half-lives of the accumulated fallout of 20, 15, 10

and 5 years, respectively. A correlation analysis showed that a half-life of 28 yr gave the best fit to the observed data (r=0.9825***).

In the case of cultivated soil, the measurements from 1973 (RRD73) and 1975 (RRD75) show that the mean ratio between the activity levels in cultivated and uncultivated soil was 0.92 $\pm$ 0.25 (1 SD) for ^{90}Sr and 0.93 $\pm$ 0.22 for ^{137}Cs. We may thus establish that although the vertical distribution of the activity in cultivated soil differed from that of uncultivated soil (cf.1.6.2.), the cumulated activity was on an average only a little lower in the cultivated soil. On the basis of the measurements of cultivated soil in 1962 (RRD62), 1973, and 1975, the "observed half-life" of ^{90}Sr (and ^{137}Cs) was estimated at 20-25 years in cultivated soils.

Figure 1.6.1. suggests that radioactive decay was mainly responsible for the reduction of the radionuclide content of the soil. However, other factors, e.g., run-off and removal by crops, may also have contributed. It was estimated (D.1.6.1.1.) that the overall decay factor of ^{90}Sr in Danish cultivated soils for the various removal processes (radioactive decay included) was: $\lambda_c = 0.036\ yr^{-1}$, during 1950-1974 and would in future approach the decay factor for ^{90}Sr, i.e. $0.025\ yr^{-1}$. The decay factor $\lambda_c = 0.036\ yr^{-1}$ during the past period, which corresponded to an "observed half-life" of 19 years, was thus compatible with actual observations in Danish cultivated soil. ERIKSSON (Er77) estimated the effective half-life of ^{137}Cs and ^{90}Sr in the root zone of Swedish soils at 23 and 22 yr, respectively.

The variability among years (table B.1.6.1.)(1962-1974) of the ^{90}Sr levels in Danish soil was 0.20. This time variability may be considered as the theoretical minimum for ^{90}Sr from global fallout in terrestrial ecosystems in Denmark, in the same way as the time variability in the precipitation and air samples was considered the theoretical maximum. The reasoning behind these assumptions was the fact that terrestrial ecosystems obtain their ^{90}Sr contents through varying amounts of direct and indirect contamination, i.e. from fresh as well as old deposits. If the contamination is mainly derived from new

deposits, the time variation resembles that of ^{90}Sr in precipitation and air; on the other hand, if the sample in question has obtained its ^{90}Sr from the cumulated debris in the soil, the time variability approaches that of ^{90}Sr in soil. However, as discussed below (2.2.1.), environmental factors other than fallout vary with time and may thus influence the time variability.

Anovas of the ^{90}Sr and ^{137}Cs data on soil showed a significant, local variation. The highest levels were found at Studsgård, Askov and Jyndevad, and the lowest at Tystofte and Ledreborg. The variation was correlated to the local precipitation pattern ($r = 0.7943^{**}$ for the 10 stations). We may therefore conclude that dry fallout either plays a minor role or that the amounts of dry fallout are proportional to the amounts of precipitation. The local variability was 0.13 (table B.1.6.1.), which was lower than that observed for precipitation (CV_p location = 0.16), because Studsgård showed lower soil than rain levels, while the data from Abed were reversed. The deviations between soil and precipitation data from these two locations were probably due to local soil characteristics and environmental conditions. In the sandy soil at Studsgård some ^{90}Sr may have penetrated below the sampling depth and the total deposit of ^{90}Sr in the soil was thus underestimated. At Abed the soil was sampled in a meadow (table A.1.1.6.2.), and some flow of ^{90}Sr from areas more highly situated may have enhanced the soil ^{90}Sr.

Studies of ^{137}Cs in soil (Ka72, Kr73, Ha74b, Ha58-78) have all shown greater $^{137}Cs/^{90}Sr$ ratios than the theoretical value of 1.45 (Harl72). The $^{137}Cs/^{90}Sr$ mean ratio in Danish soil lay between 1.6 and 1.7, i.e. not significantly different from the mean ratio found in air samples collected at Risø since 1962. The ratio higher than the theoretical 1.45 may be a result of fractionation (Ed59, Fr61). MAMURO et al. (Ma65) thus observed that the highly radioactive primary fallout particles collected in the autumns of 1961 and 1962 after the Russian test series were impoverished in ^{137}Cs. This was compatible with the delayed, long-range global fallout being enriched in ^{137}Cs. Assuming that the debris over Denmark contained relatively large amounts of global fallout as compared with primary

fallout, because Denmark is distant from the test sites, we may expect a relatively larger ^{137}Cs content in the air and soil samples.

Soil samples have not been collected routinely from the Faroes and Greenland. From the fallout data (1.3.) the accumulated ^{90}Sr in 1967 at Thorshavn was estimated at 78 mCi km^{-2} and in Godthåb in 1970 at 42 mCi km^{-2}; HARDY et al. (Hard72) found 9 mCi ^{90}Sr km^{-2} in 1970 at Thule Air Base, which is a few kilometers south of Dundas. These figures are all in reasonable agreement with the ^{90}Sr levels shown in table D.1.6.1.2. As regards Tværå, there is no information on rainfall and fallout from this location. The soil data from Tværå suggested that the fallout rate was between those at Thorshavn and at Klaksvig. The ^{137}Cs concentrations of the Faroese and Greenlandic soil samples were in general higher than the expected 1.6 times the ^{90}Sr concentrations. As also observed in the case of Danish soils (1.6.2.), the "median depths" of ^{137}Cs in the Faroese and Greenlandic soils were lower than that of ^{90}Sr.

1.6.2. The vertical distribution of radioactive debris in soils

Several investigators (Th60, Sc60, An66) have studied the vertical penetration of radionuclides in soil and have described the distribution and migration by mathematical expressions. The models have, however, been unable to describe the long-term situation under natural conditions in the environment. As mentioned above, the environmental factors that determine the retention and distribution of radionuclides in the soil are rather complex, and it seems difficult to make allowances for all significant factors in theoretical models.

Over a period of 8 years SQUIRE (Sq66a, Sq66b) studied the behaviour of ^{90}Sr and ^{137}Cs in soil. These studies were carried out with artificially added activities applied at one time to experimental concrete cylinders. SQUIRE observed that the median depth of the activity was proportional to the square root of time since deposition of the activity; this she found "compatible with movement being determined largely by the random diffusion of ions in the exchange complex of the soil".

As the ^{90}Sr fallout in Danish soils had penetrated by 1975 to a depth of approx. 50 cm (RRD75), and as fallout commenced approx. 25 years ago, it was assumed that the front of the activity deposited t years ago had reached a depth D cm:

$$D = 10\sqrt{t}. \qquad \text{(Eq.1.6.2.)}$$

Eight years after the application of ^{90}Sr to 3 soils of sandy loam and one of calcareous loam, SQUIRE (Sq66a) observed that the activity front had reached a depth of 20-30 cm. Equation 1.6.2. predicts a depth of 28 cm.

The distribution of the activity of the deposit of a given year may be described by:

$$Y = \frac{1 - e^{\frac{-ax}{D}}}{1 - e^{-a}} \qquad \text{(Eq.1.6.3.)}$$

where $\underline{Y}$ is the fraction of the total integrated activity in the soil column present down to a depth $\underline{x}$ cm ($x \leq D$) and $\underline{a}$ is the "vertical distribution coefficient". This model was derived from experimental studies of the ^{137}Cs activity profiles in British soils (Ga63), which suggested vertical distributions compatible with Eq.1.6.3. The "vertical distribution coefficient" depended on environmental factors, e.g., soil texture, and on the radionuclide considered. For ^{90}Sr in sandy soils, a equalled approx. 3 and in loamy soils approx. 1. The variability of ^{90}Sr among depths was in fact higher for loamy than for sandy soils (table B.1.6.1.). As an average for the Danish soils, a = 2 fitted the data with good approximation (fig.1.6.2.). A comparison between the experimental data of SQUIRE (Sq66a) and this model showed that the UK experiments yielded a-values of 3.5 and 2 for soils without and with application of fertilizers, respectively. The Danish average value thus corresponded to the UK value for fertilized soil.

A high a-value implies a shallow median depth, i.e. the bulk of the activity remains in the uppermost layers of the soil; the distribution is uneven. The opposite is the case if the a-value is small, the activity is then more evenly distributed

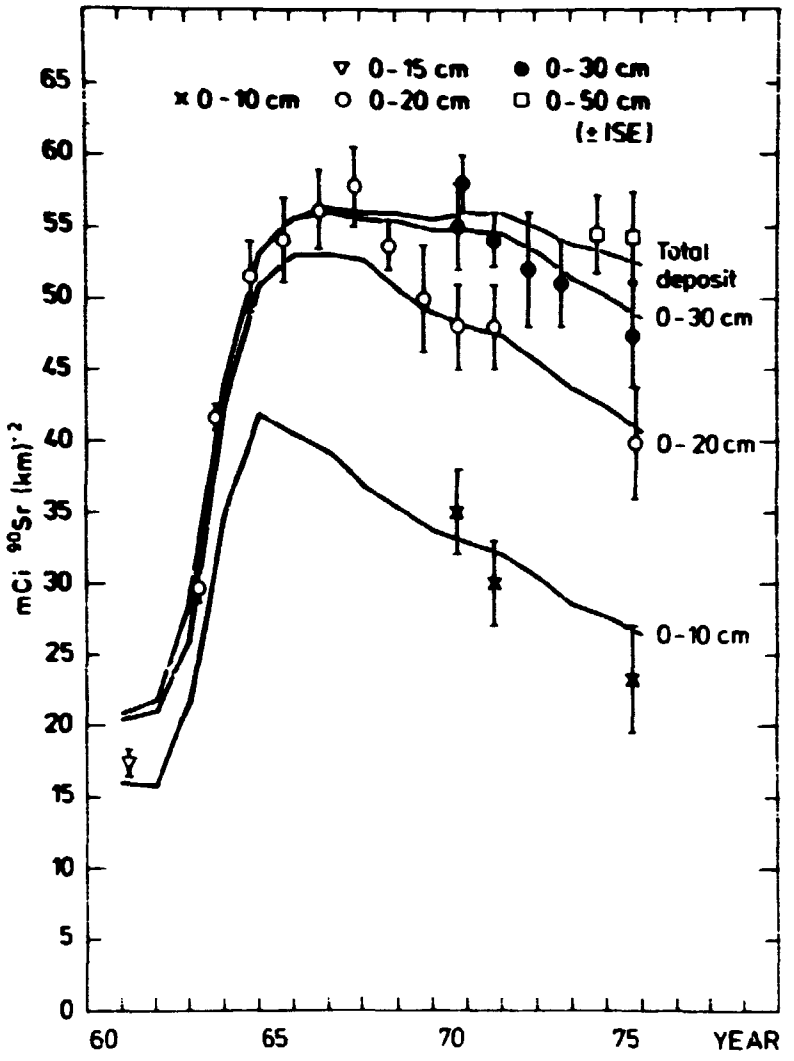

Fig. 1.6.2. The integrated ^{90}Sr levels in uncultivated Danish soil measured at various depths compared with the calculated levels (the curves). The total deposit was calculated from Eq. 1.6.1. applying an effective half-life of 28 yr (cf. Fig. 1.6.1.). The vertical distribution was calculated from Eqs. 1.6.2. and 1.6.3 applying a vertical distribution coefficient a = 2 (cf. the text). Eq. 1.6.3. implies that the maximumconcentrations will remain in the uppermost soil layer. Actually the maximum is displaced downward with time. However as shown the displacement is so slow, that it was not detectable, when 10 cm thick samples were analysed. The samples were collected at the state experimental farms (Fig. A.1.1.3.1.) and in 1970 at an additional ten locations distributed over the country (cf. Fig. A.1.1.6 and Table A.1.1.6.1.).

in the soil column. If Y = 0.5 is inserted into Eq.1.6.3. and the equation is solved with respect to x, the "median depth" $x=a_m$ becomes:

$$a_m = \frac{- D \cdot \ln [0.5 \ (1 + e^{-a})]}{a} \ . \qquad \text{(Eq.1.6.4.)}$$

The root uptake of ^{90}Sr by the crops is influenced by the median depth of the activity in the soil. As shown in D.1.6.2., the effective halflife for root uptake of ^{90}Sr may be estimated for various crops in various soils. According to these estimates, both cultivated and uncultivated Danish soils showed effective halflives of ^{90}Sr of the order of 15 years, somewhat lower for crops with a shallow root system such as grass and a little higher for crops with deeper roots such as cereals and root crops. If fresh fallout ceases, we can in the long run expect increasing effective half-lives for root uptake of ^{90}Sr because the half-lives observed in the soil will increase (1.6.1.), although at first we may expect a temporary decrease in the effective half-life for cultivated soils.

Several experiments (Ru66b) provide convincing evidence that the conversion of ^{90}Sr into sparingly soluble forms is unlikely to reduce its availability to plants to a significant degree over a period of several years. However, whether or not such a reduction in availability could take place over long periods has not yet been clarified.

If Eq.1.6.3. is applied to ^{137}Cs, the "vertical distribution coefficient" is close to 5 for Danish average soils. The higher a-value for ^{137}Cs than for ^{90}Sr was tantamount to a steeper gradient of the vertical ^{137}Cs distribution in the soil, in agreement with the well known (Fr66) higher retention of ^{137}Cs than of ^{90}Sr by clay minerals.

1.6.3. Sediments

Sediments and sedimentary particles remove radionuclides from water. Certain radionuclides are more strongly sorbed on sediment particles than others. More ^{137}Cs is thus carried by sediments than ^{90}Sr (cf.1.5.1.). Nuclear debris moves through the sediments after deposition. Diffusion through interstitial water is one way of transportation. Where sediment accumulation is rapid, the radioactive particles may be covered by later sedimentary deposits before diffusion, and other transport processes can displace the nuclides. In such cases it may be possible to identify single years of deposition in the sediment

layers and thus to estimate the sedimentation rate (Pe73). Among the other transport processes of radionuclides in sediments the burrowing of benthic organisms, currents near the bottom and ice movements may play a role.

Sediment cores were collected in inner Danish waters in 1975 (RRD75) and 1976 (RRD76) and analyzed for ^{137}Cs. The mean half-depth of the 11 locations studied was 4 cm, which is comparable to data from the Irish Sea (He75) showing half-depths between 4.1 and 7.2 cm. The integrated mean level in sediments from inner Danish waters was estimated at 14 $\pm$ 6 mCi ^{137}Cs km^{-2} (1 SD), or approx. 15% of the ^{137}Cs fallout found in soil. This estimate was approx. half that obtained from a comparison of the transfer coefficients for ^{90}Sr and ^{137}Cs in inner Danish waters (cf.1.5.1.). The sediment samples may, however, not be representative of inner Danish waters as a whole because most of them were collected along the Swedish west coast.

In November 1973 and in April 1974 sediment core samples were collected to a depth of 20 cm from lake Kongsø in central Jutland by the Laboratory of Freshwater Biology in Hillerød. The samples were analyzed for ^{137}Cs in 5 mm sections, and the activity decreased exponentially with a half-depth of 4 cm, i.e. equal to that observed for sea-water locations. The integrated mean level of the two samplings from Kongsø was 79 $\pm$ 15 mCi ^{137}Cs km^{-2}. This deposition came close to the theoretical amount of ^{137}Cs in soil from fallout ($\sim$ 90 mCi ^{137}Cs km^{-2}), and it was definitely higher than the inventory found in sea-water sediments. The distribution coefficient, K_d, between ^{137}Cs in sediments and lake-water was estimated at 10 pCi g^{-1}/0.6 $\cdot$ 10^{-3} pCi ml^{-1} $\sim$ 2 $\cdot$ 10^{4} ml g^{-1}. The K_d value for ^{137}Cs in Danish sea-water was (10^{2} - 10^{3}) ml g^{-1}, which was in agreement with experimental observations (Du71). The distribution coefficient K_d of ^{137}Cs for sediments in fresh water was thus more than an order of magnitude greater than the K_d for sea-water sediments.

Several investigators have observed a close relationship between the vertical distribution of ^{137}Cs and $^{239,240}Pu$ in sediments (He75, No72, Ed75). In a study of sediments from inner Danish

waters (RRD76), the $^{239,240}Pu/^{137}Cs$ ratios in the various sediment layers suggested that the vertical distributions of the radionuclides were similar. The integrated plutonium deposit corresponded to the levels found in Danish soils (RRD76), i.e., about 1.5 mCi $^{239,240}Pu\ km^{-2}$.

The studies of $^{239,240}Pu$ at Thule (V) have shown with approximation that plutonium from a single release was distributed exponentially both horizontally and vertically. The half-depth value was 1-2 cm, and the K_d value was $10^5 - 10^7$ ml g^{-1}. This indicated that nearly all plutonium was found in the sediments, which is in agreement with other observations (He75). The investigations also suggested that biological transport may be important for the vertical displacement of plutonium in sea sediments.

Strontium-90 has not been studied in Danish sediments but various investigations (Du71) have shown K_d values of the order of $(10 - 10^2)$ ml g^{-1} in marine sediments and 120-440 in lake sediments (Le72), i.e. more than an order of magnitude less than that observed for ^{137}Cs. We may therefore neglect the ^{90}Sr inventory of sediments compared to the amounts present in sea and lake water.

1.7. Conclusions

1.7.1. General

The atmospheric nuclear test explosions performed since the detonation of the first nuclear weapon in 1945 have contaminated air, water and soil with a variety of radionuclides. The residence time in the environment of the majority of these nuclides has been relatively short, but a few remain even decades after their creation. Among these are ^{90}Sr and ^{137}Cs which, due to their chemical affinity with the biologically important elements calcium and potassium, respectively, are readily transferred from the abiotic environment to living tissues.

The measurements of the ^{90}Sr and ^{137}Cs concentrations in air, water and soil constitute the basis of the prediction models estimated for the various samples in this study, and hence for the calculation of the transfer factors from fallout to man.

The variabilities of the abiotic environmental samples are useful as indicators of the routes of contamination of the biological environmental samples.

1.7.2. Air

The air concentrations of ^{90}Sr and ^{137}Cs showed similar variabilities among years ($CV_{p\ year} = 1.7$) as well as among months within the various years ($CV_{p\ months} = 0.6$). The mean $^{137}Cs/^{90}Sr$ ratio was approx. 1.6 and showed neither local nor yearly variations.

An empirical model was proposed for the estimation of transfer factors from injection to ground-level air concentrations (fCi m^{-3} y per MCi globally injected by the test explosion); according to this model, the transfer factor of a given radionuclide was related to the effective half-life in the atmosphere of the nuclide by a power function. The transfer factor for ^{90}Sr and ^{137}Cs was estimated at 8 fCi m^{-3} y per MCi.

The total integrated air levels originating from nuclear weapons testing was estimated at 0.12 pCi $^{90}Sr\ m^{-3}$ y ($4.4 \cdot 10^{-3}$ Bq m^{-3} y), 0.22 pCi $^{137}Cs\ m^{-3}$ ($8.1 \cdot 10^{-3}$ Bq m^{-3} y) and 2.9 fCi $^{239,240}Pu\ m^{-3}$ y ($1.07 \cdot 10^{-4}$ Bq m^{-3} y). A "standard man" breathes 7.300 m^3 air per year (Ic59), hence 0.88 nCi ^{90}Sr, 1.6 nCi ^{137}Cs and 21 pCi $^{239,240}Pu$ from nuclear weapons debris have been inhaled under Danish conditions by standard man. As regards ^{90}Sr and ^{137}Cs, these intakes were 4 and 2.5‰, respectively, of the corresponding intakes with the diet (cf.4.2). From the ICRP Task Group Lung Model (Ic66), assuming a mean fallout particle size of 0.4μ (Sh66), it was estimated that 9% of the ^{90}Sr inhaled was absorbed into the blood, 45% was carried to the GI tract, while 46% was expirated. The corresponding values for ^{137}Cs were 44%, 9% and 47%, respectively. It was thus evident that the contributions to body burden from the inha-

lation of ^{90}Sr and ^{137}Cs were negligible as compared with intakes from diet (cf.4.7). In the case of plutonium, uptake from the diet is immaterial (Un77, IX) and inhalation is the important pathway. BENNETT estimated the plutonium doses to man (Be74). He found that the cumulative dose to bone through the year 2000 was 1.5 mrad from 5.8 fCi $^{239,240}Pu\ m^{-3}\ y$, hence the bone dose under Danish conditions to standard man was estimated at 0.8 mrad (or 17 mrem), and the lung dose became 0.4 mrad (or 8 mrem); the liver received a dose similar to that to the lungs according to BENNETT.

1.7.3. Precipitation

The ^{90}Sr concentration in precipitation showed a variability among years and among months similar to that in the air concentrations of ^{90}Sr. Furthermore, the annual variabilities of ^{90}Sr in Faroese and Greenlandic precipitation corresponded to those in Danish rain water, and the annual depositions of ^{90}Sr in the Faroes and Greenland were proportional to the Danish levels. An annual deposition of 1 mCi $^{90}Sr\ km^{-2}$ in Denmark thus corresponded to 2.08 mCi km^{-2} in the Faroes and 0.75 mCi km^{-2} in West Greenland (Godthåb district). There was no significant variation among locations as regarded pCi $^{90}Sr\ l^{-1}$ rain in Denmark, but the deposition (mCi $^{90}Sr\ km^{-2}$) was proportional to rainfall, hence the local variability of depositions ($CV_{p\ locations} = 0.16$) was similar to that of mm precipitation.

The deposition velocity of global ^{90}Sr dry fallout in Denmark was $2 \cdot 10^{-3}\ m\ s^{-1}$ and the observed washout ratio (or scavenging ratio) for ^{90}Sr was 0.99 pCi l^{-1} $(fCi\ m^{-3})^{-1}$. On the average, the ^{90}Sr concentration in rain water (pCi l^{-1}) has thus shown the same numerical value as the air concentration in fCi m^{-3}. The coefficient of variation of the monthly scavenging ratios was 0.5.

The total deposition of ^{90}Sr in Denmark from nuclear weapons debris was 73 mCi km^{-2} ($2.7 \cdot 10^{9}$ Bq km^{-2}) with 81.3 mCi km^{2} ($3.0 \cdot 10^{9}$ Bq km^{-2}) in Jutland and 64.8 mCi km^{-2} ($2.4 \cdot 10^{9}$ Bq km^{-2}) in the Islands. The corresponding ^{137}Cs levels were 1.6 times higher. The total ^{90}Sr deposition in the Faroes was 152 mCi km^{-2} ($5.6 \cdot 10^{9}$ Bq km^{-2}), and in West and East Greenland

the levels were 55 and 34 mCi $^{90}Sr\ km^{-2}$, respectively, ($2.0 \cdot 10^{9}$ and $1.26 \cdot 10^{9}$ Bq km^{-2}).

1.7.4. Fresh water

The Danish ground water showed a lower variability among years of the ^{90}Sr concentrations ($CV_{p\ years} = 0.6$) than rain water. This results from the dependence of the ground-water concentrations upon the ^{90}Sr accumulated in the soil layers. On the other hand, the local variability ($CV_{p\ locations} = 0.5$) was higher for ground-water than for precipitation because the soil characteristics influenced the ground-water concentrations. For similar reasons, stream water as well as lake water also showed higher local variabilities than the rain.

The radioecological sensitivities of Danish ground, stream and lake water were $7 \cdot 10^{-3}$, 0.3 and 1 pCi $^{90}Sr\ l^{-1}$ y per mCi $^{90}Sr\ km^{-2}$, respectively. One ground water station with sand as a filtering layer showed a sensitivity that was two orders of magnitude higher than the other ground-water borings.

The total run-off in Denmark from the land area to the sea was 4.6 mCi $^{90}Sr\ km^{-2}$ up until 1974. The annual run-off has, on the average, been 5% of the deposited ^{90}Sr; in recent years it has decreased to approx. 2%.

Because of the low ^{90}Sr concentrations in ground water, the intake of ^{90}Sr with drinking water was 5 pCi $(cap)^{-1}$ per mCi $^{90}Sr\ km^{-2}$, or 2% of the intake of ^{90}Sr with the diet (table 4.2.2.). The ^{137}Cs concentration in Danish drinking water is even lower than the ^{90}Sr level because the Danish soil retains ^{137}Cs more efficiently than ^{90}Sr (1.6).

Although drinking water is unimportant as a source of ^{90}Sr in Denmark, it is a main contributor to the human intake of stable Sr, as nearly half of the stable Sr (200-300 mg $(cap)^{-1}$ y^{-1}) is derived from drinking water. The local variations of stable Sr were pronounced. Water from Lolland-Falster thus contained 12 times more Sr than water from West Jutland.

Drinking water in the Faroes was derived from surface waters. The radioecological sensitivity was 0.1 pCi $^{90}Sr\ l^{-1}$ y per mCi $^{90}Sr\ km^{-2}$; the variability among years was 0.6.

In Greenland the drinking water showed a high local variability ($CV_{p\ locations}$ = 1.1) because the origin of the water at the various locations differed. At some Greenland locations, old ice free of radioactive fallout was used, at others the water was fresh rain water with relatively high radionuclide concentrations. The radioecological sensitivity of drinking water from West Greenland was 0.5 pCi $^{90}Sr\ l^{-1}$ per mCi $^{90}Sr\ km^{-2}$.

1.7.5. Sea water

The ^{90}Sr concentrations in inner Danish waters were inversely related to the salinity during 1966-1972, because the highest levels came from the little saline Baltic water, while highly saline North Sea water was low in activity due to dilution from the Atlantic Ocean. Since 1972 releases, especially of ^{137}Cs, from Windscale have been increasingly detectable in inner Danish waters and the activity levels have become directly related to salinity.

The variability in the ^{90}Sr levels among years was low ($CV_{p\ years}$ = 0.2) as compared with air and precipitation, but it corresponded to that observed in soil.

The radioecological sensitivity of Danish waters (salinity 16‰) was 0.4 pCi $^{90}Sr\ l^{-1}$ y per mCi $^{90}Sr\ km^{-2}$. In comparison, North Atlantic surface water showed a sensitivity of 0.03. Greenland coastal waters showed sensitivities similar to those observed in Danish waters.

The variability of ^{90}Sr in North Atlantic sea water from the Faroes and Greenland was higher ($CV_{p\ years}$ = 0.5) than in Danish waters due to the more rapid dilution of the deposited debris in a deep ocean than in shallow waters.

Deep sea samples from the North Atlantic suggested a deposition of approx. 150 mCi $^{90}Sr\ km^{-2}$ over this area of the sea. This

observation is compatible with the deposition of ^{90}Sr in the Faroes, but twice as high as the estimate from North Atlantic weather ships.

1.7.6. Soil and sediment

The effective half-life of ^{90}Sr in uncultivated Danish soil was not significantly different from the radiological half-life of 28 years. Hence the soil measurements suggested that the deposition of ^{90}Sr estimated from rain-funnel sampling (1.3.) corresponded to the deposition in the pastures used for soil sampling. The variability of ^{90}Sr in Danish soils among years was 0.2 and the local variability was 0.13, which was close tc that of rain fall.

As in air samples, the $^{137}Cs/^{90}Sr$ ratio in Danish soils was in the range 1.6-1.7. The median depth of ^{137}Cs was lower than that of ^{90}Sr, because ^{137}Cs is more easily retained in the soil layers. Hence $^{137}Cs/^{90}Sr$ decreased with increasing sampling depth.

The vertical penetration of ^{90}Sr in soil was proportional to the square root of years elapsed since the deposition took place. An empirical model for the median depth of ^{90}Sr was combined with a model for the relative root uptake ^{90}Sr by Danish crops. The model suggested that the effective half-life of ^{90}Sr in crops grown in Danish soil would be approx. 15 years, somewhat lower for grass but higher for cereals and vegetables. In the long run, the effective half-life in the crops would approach the radiological half-life.

Sea sediments from inner Danish waters contained 14 $\pm$ 6 mCi ($\pm$1SD) ^{137}Cs km^{-2} ($5.2 \cdot 10^{8}$ Bq km^{-2}) and 1.5 mCi $^{239,240}Pu$ km^{-2} ($5.6 \cdot 10^{7}$ Bq km^{-2}). The plutonium level equalled that in soils, while the ^{137}Cs figure was approx. 15% of the ^{137}Cs deposit. A lake sediment sample contained nearly all the ^{137}Cs expected in a corresponding soil sample taken from the same area, suggesting that the distribution factor between water and sediments is an order of magnitude higher for lake sediments compared with sea sediments from inner Danish waters.

2. GRAIN AND OTHER VEGETABLE PRODUCTS (the producers)

2.1. Introduction

In general, plants are the primary recipients of radioactive contamination to the food chain from the abiotic environment. Vegetation may be subject to direct and indirect contamination (Ru66a). In the case of terrestrial vegetation, direct contamination implies an uptake of radioactive debris from the atmosphere by the above-ground parts of plants. Indirect contamination is a sorption of debris from the soil by the root system of plants. Aquatic plants are contaminated by contact with radioactive water and sediments (Lo71). Sea weed on the surface may receive direct contamination from the atmosphere.

Whereas all radionuclides may be taken up by vegetation as a result of direct contamination, relatively few are of relevance as regards root uptake. Among these, ^{90}Sr is the most important, because it continues to enter the food chain many years after its deposition; and this is the main reason why ^{90}Sr is of foremost interest in radioecology. The radionuclides that occur both in a soluble form and also translocate within the plants are of special interest as direct contaminants, because they may concentrate in parts of the plants which have not themselves been exposed to direct contamination. This feature may be very important for the radionuclide concentrations in cereal grain, and thus it has been examined in a number of field experiments (VI, VII, VIII, Aa72b) described in the present study.

Vegetable products are an important constituent of the human diet; in Denmark the daily per capita consumption is approx. 0.7 kg, which equals the consumption of animal products. Some products are used directly for human consumption, such as cereal grains, vegetables, and fruit. But plants may also play an indirect part in the intake of radionuclides from foods. The concentration of radionuclides in grass and beets thus plays

an important role in the radionuclide content of cows' milk, while the presence of radioactive debris in a vegetation cover, such as lichen, strongly influences the radioactive contamination of reindeer meat.

2.2. Cereal grain

Der var saa deiligt ude på landet; det var Sommer! Kornet stod gult, Havren grøn.
DEN GRIMME ÆLLING

Summertime! How lovely it was out in the country, with the wheat standing yellow, the oats green.
THE UGLY DUCKLING

Studies of fallout nuclides in the Danish diet have revealed that cereal products, especially rye bread, play an essential part in the radioactive contamination of the diet. When grain is contaminated by radioactive fallout, the importance of direct contamination is greater than that of the indirect. The outer parts of the grain therefore carry the major share of the contamination. Whole-grain flour (100% extraction) consequently shows higher concentrations of fallout radioactivity than flour made of the inner parts of the grain only. Hence, rye bread made of 100% extraction flour contains more radioactivity than, e.g., white bread of 70% extraction flour. As the preference in most western countries is for white bread rather than dark rye bread, grain products are relatively more important for the radioactivity content of the Danish diet, which does contain rye bread, than for the diet elswhere. The present study has therefore emphasized cereals.

The first UNSCEAR report (Un58) summarized the earliest measurements of ^{90}Sr in grain. The samples were rice and wheat collected in Japan, the USSR and the USA in 1956. The first systematic study of ^{90}Sr in rye, barley, wheat and oats was carried out in 1959 in Denmark (RRD59). Extensive studies of ^{137}Cs as well as ^{90}Sr in wheat and in milled wheat were performed in Canada on samples from the crop years 1957-1959 (Gr60). Since 1958 UNSCEAR has occasionally (Un62, Un69) published summaries of worldwide studies of fallout nuclides in grain and grain products.

2.2.1. Variation with time, species and location

Radionuclide concentrations in cereal grain have varied with time, species and location as appears from figs. 2.2.1.1-2.2.1.4 (cf. also tables B.2.2.1.-B.2.2.5.); the time variation was the most pronounced (tables B.2.2.7.-B.2.2.8.). The variability

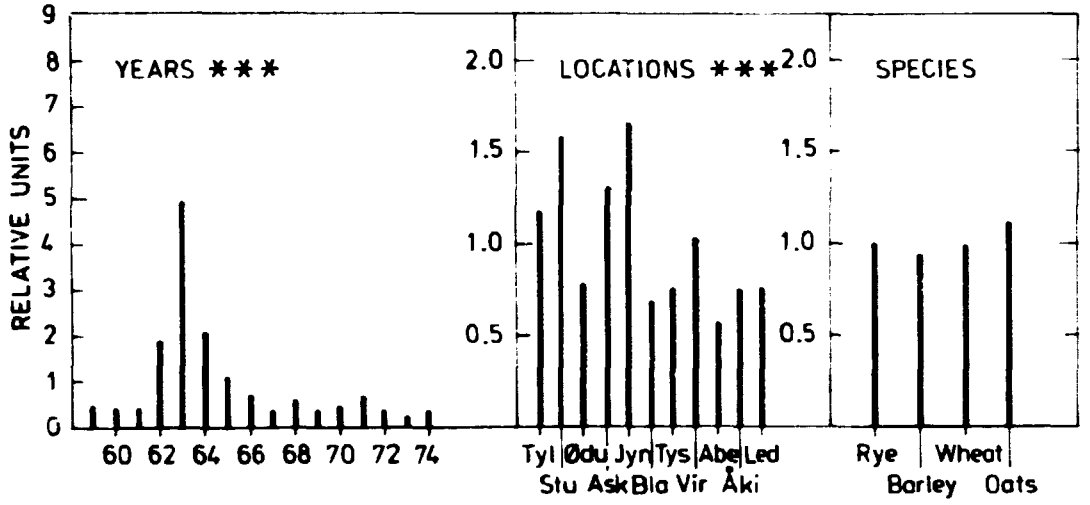

Fig. 2.2.1.1. The variation of pCi ^{90}Sr kg^{-1} in Danish cereal grain among years, locations (fig. A.1.1.3.1.) and species. The bars are the concentrations relative to the grand mean 77 pCi kg^{-1} (= 1 at the relative scales).

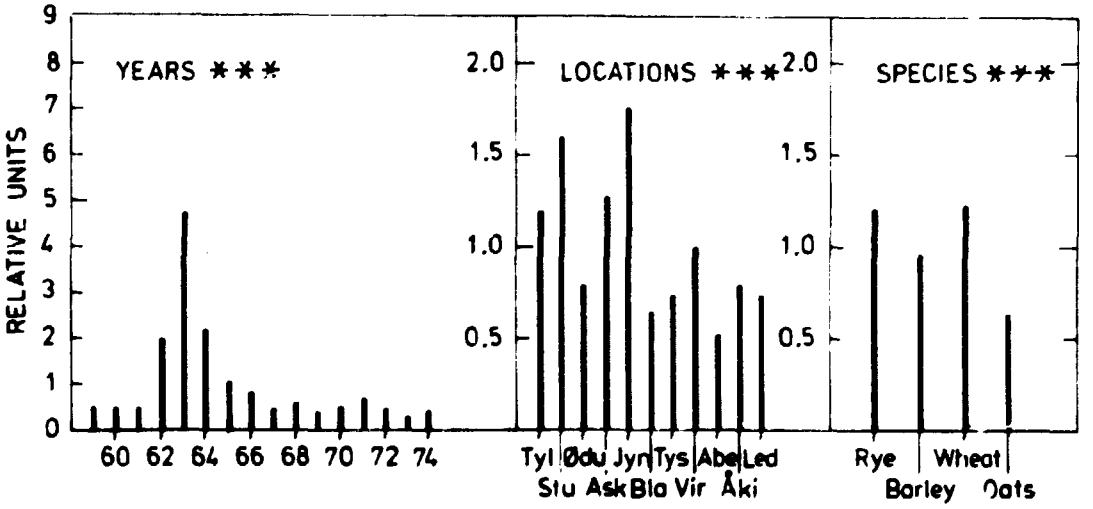

Fig. 2.2.1.2. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in Danish cereal grain among years, locations (fig. A.1.1.3.1.) and species. The bars are the concentrations relative to the grand mean 155 pCi (g Ca)$^{-1}$ (= 1 at the relative scales), (cf. remarks to table 2.2.1.1.).

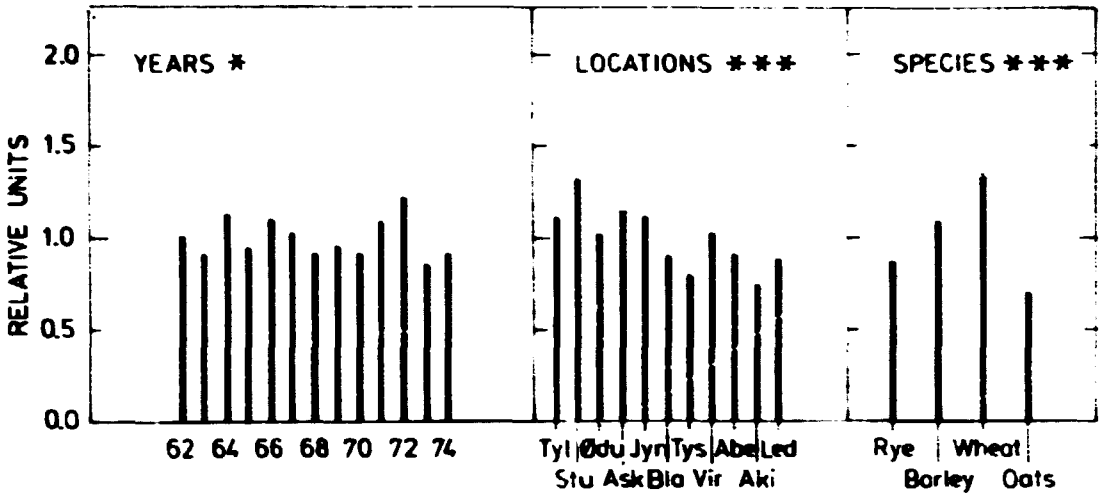

Fig. 2.2.1.3. The variation of stable Sr in Danish cereal grain among years, locations (fig. A.1.1.3.1.) and species. The bars are the levels relative to the grand mean 2.8 mg (g Ca)$^{-1}$ (= 1 at the relative scale).

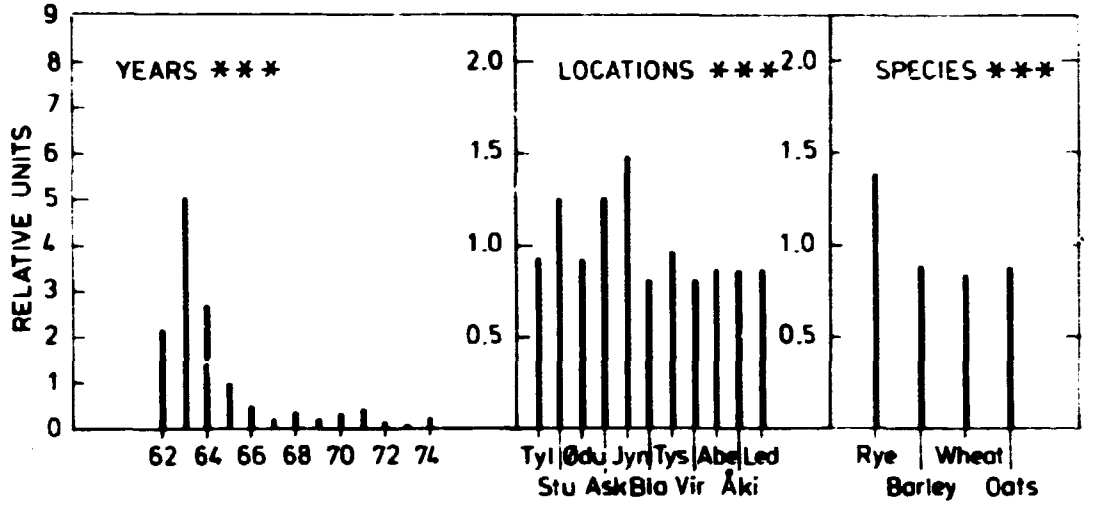

Fig. 2.2.1.4. The variation of pCi ^{137}Cs kg^{-1} in Danish cereal grain among years, locations (fig. A.1.1.3.1.) and species. The bars are the concentrations relative to the grand mean 192 pCi kg^{-1} (= 1 at the relative scales).

among years from 1959-1974 was 0.85 for ^{90}Sr. In the case of ^{137}Cs it was even higher; from 1962-1974 it was thus 1.77 or nearly equal to the variability of precipitation observed in that period (1.3.). The time variability of ^{137}Cs was greater than for ^{90}Sr because there is no moderating effect from root uptake of ^{137}Cs from the soil in Denmark. Rye and barley showed higher variability among years than wheat and oats because the

former two species are more subject to direct contamination than the latter two; this is partly due to morphological reasons (greater surface of the ears because they have awns), and partly to physiological reasons, viz., a greater root uptake of ^{90}Sr by wheat and oats. The various parts of the country showed the same time variability. As expected, the variability among years was highest in the high-fallout period 1962-1966. Although the fallout rate and the accumulated fallout are the factors chiefly responsible for the time variation of ^{90}Sr (and ^{137}Cs) in grain, other factors may also be of significance (Aa72a), such as variation in grain yields from year to year, increasing air pollution, alterations in agricultural practice, e.g. switching from using a reaper to using a combine harvester, and growing different varieties of grain (D.2.2.1.).

As appears from table B.2.2.9. the variability among species was higher for Pu ($CV_{p\ species} = 0.73$) than for any of the other nuclides studied, i.e. ^{90}Sr, ^{137}Cs and ^{54}Mn. This can be ascribed to a combination of the morphology of the cereal crops and the low solubility of Pu (IX). Strontium-90 showed a lower variability among species than ^{137}Cs, because the high susceptibility of rye and barley to direct contamination with ^{90}Sr was counteracted by a relatively high root uptake of ^{90}Sr by wheat and oats. In 1962-1966 the variability of ^{90}Sr among species was lower in Jutland than in the Islands; during 1967-1972 the opposite was the case. The reason was that during the high fallout period in 1962-1966, when direct contamination prevailed, this domination was more pronounced in the soils with high Ca of the Islands than in the more sandy soils of Jutland. This resulted in a levelling of the ^{90}Sr concentrations in the species from Jutland because, as mentioned above, wheat and oats are more sensitive to indirect contamination than rye and barley. During 1967-1972 when direct contamination was less dominant, the levelling effect of the remaining direct contamination was most effective in the Islands, because the indirect contamination of wheat and oats in Jutland clearly prevailed at that time. In the case of ^{137}Cs, as expected, no evident trend was found between the variability among species from Jutland and the Islands because direct contamination dominated the uptake of ^{137}Cs.

Before 1966, rye showed both highest ^{90}Sr as well as highest ^{137}Cs concentrations (Aa66c). Throughout the entire period of observation, the ^{137}Cs concentration in rye was approx. 50 per cent higher than in the other three species, which did not differ significantly from one another. Since 1967, when direct contamination became less important in case of ^{90}Sr, oats have shown the highest and rye the lowest pCi ^{90}Sr kg^{-1} figures. There may be several reasons why rye is more susceptible to direct contamination than the other species; the two main ones being the morphology and the precocity of rye. Rye grows taller than the other species. The straw length is 120-130 cm, compared with 100-120 cm for wheat (Pe63) and even less for barley and oats. The yield of rye is generally lower than of the other species (Pe63, Lu71). Furthermore, the grain of rye is relatively small in size (Pe63) and the surface-to-volume ratio consequently greater than for the other species. Finally, rye carries awns whereas other Danish cereal crops, except barley, are normally without awns. The ears of rye emerge by the end of May, while the other species ear by the end of June. The precocity of rye is of special relevance for ^{90}Sr because the important period for the direct contamination with this and other nuclides not translocated within the plant is after ear emergence (Mi59, VI, VIII).

Two important factors influence the local variation: the amount of precipitation (fallout) and the agricultural conditions (soil type, cultivation practice, etc.). In Jutland, the amounts of precipitation, and hence of fallout, are approx. 20 per cent higher than in eastern Denmark (table A.1.1.6.2.). Furthermore, the soils of the experimental farms in Jutland are on the average less fertile than those of farms in the east, and the Ca content is lower. All these factors enhance the contamination of grain with fallout ^{90}Sr (Cham70, Aa72a, An67a), especially at the sandy locations in Jutland (Studsgård and St. Jyndevad). The stable Sr to Ca ratio is higher in samples from the sandy stations, as illustrated in fig.D.2.2.1., which indicates a relatively higher root uptake of Sr (Aa66c). As appears from tables B.2.2.7. and B.2.2.8., the local variability was more pronounced for ^{90}Sr than for ^{137}Cs (and for ^{54}Mn and $^{239,240}Pu$ as well (table B.2.2.9.)). The relatively higher root uptake of ^{90}Sr in Jutland accentuated the local variations. In

the case of ^{90}Sr, the local variability of rye was lower than that of the other species because the ^{90}Sr concentration in rye depends relatively more on direct contamination than does the concentration in other species, which means that the amplifying effect on the local variability of root uptake is less evident in rye. The local variability among the four stations in Jutland: TYL, STU, ASK and JYN, was generally higher than the local variability among the stations in the Islands: BLA, TYS, ABE, LED and AKI. This was ascribed to differences in fallout pattern as well as to variations in soil characteristics and agricultural practice. In the case of ^{137}Cs, the local variability was close to that observed for precipitation (mCi ^{90}Sr km^{-2}, cf.1.3.) suggesting than the cereal grain was contaminated by direct contamination only. During the low fallout period 1967-1972 the local variability of ^{90}Sr was higher than in the high fallout period 1962-1966, because of the amplifying effect of indirect contamination when the importance of direct contamination decreased. As a whole, the local variability of ^{90}Sr increased throughout the period from approx. 0.3 in 1963-1965 to 0.6 in 1975-1976. In the case of ^{137}Cs the variability among locations was nearly constant until 1972, but in recent years it has shown an increase, which may indicate slight root uptake at the sandy locations in Jutland.

Table 2.2.1. Strontium-90 (pCi ^{90}Sr (g Ca)$^{-1}$) in rye and wheat collected in Jutland and the Islands in 1963 and 1973.

Year	Species	Jutland	Islands	Jutland / Islands
1963	rye	1357	953	1.42
	wheat	993	748	1.33
1973	rye	86	40	2.15
	wheat	88	34	2.59
1963 / 1973	rye	15.8	23.8	
	wheat	11.3	22.0	
1963	rye/wheat	1.37	1.27	
1973		0.98	1.18	

The anovas of the grain data revealed significant interactions between time and species, between species and location, and between location and time. Table 2.2.1. illustrates the interactions for pCi ^{90}Sr (g Ca)$^{-1}$. The years 1963 and 1973 were selected because the fallout rate as well as the accumulated fallout differed markedly between these years. In 1963 the fallout rate was high and the accumulated fallout relatively low, whereas in 1973 the fallout rate was low and the accumulated fallout relatively high. The interaction between years and locations appeared as lower activity ratios between Jutland and the Islands in 1963 than in 1973, due to the greater importance of root uptake in 1973. The interaction between species and years was manifested as higher activity ratios between rye and wheat in 1963 than in 1973, which is explained by the fact that root uptake of ^{90}Sr was relatively more important for wheat than for rye. The interaction between locations and species manifested itself as higher ratios between the activities from the two years for rye than for wheat, because the direct contamination played a greater role for rye than for wheat, which was less susceptible to the fallout rate because of the stronger influence from indirect contamination. Finally, the table also displays the second-order interaction between years, locations and species: The activity ratio between rye and wheat decreased more in Jutland than in the Islands from 1963 to 1973 because the relatively higher root uptake in 1973 influenced wheat more than rye, and because root uptake played a greater role in Jutland than in the Islands. It thus appears that the main reason for the interactions observed for ^{90}Sr in Danish cereal grain is variations in the relative importance of direct and indirect contamination. These variations are influenced by fallout rate, accumulated fallout, soil type, fertilizers (An71), grain varieties, methods of harvesting, climate, and other environmental factors.

As indirect contamination only plays a role in the case of ^{90}Sr, it was to be expected that the interactions for ^{137}Cs in grain were less evident. In fact the anovas showed that these interactions were only significant in a few cases (cf.tables B.2.2.3. and B.2.2.4.).

2.2.2. Prediction models

Tables C.2.2.1. - C.2.2.3. show the prediction models for ^{90}Sr in cereal grain. Estimates from previous models were compared with the present ones in D.2.2.2.1. The radioecological sensitivity of grain in Jutland was higher than that in the Islands for ^{90}Sr as well as for ^{137}Cs (tables C.2.2.4. - C.2.2.6.), although, as expected, it was less pronounced for ^{137}Cs because of the lack of root uptake (cf.2.2.1.). The radioecological sensitivity of cereal grain for the entire country may either be estimated directly, as shown in tables C.2.2.3. and C.2.2.6., or it may be calculated from the figures found for Jutland and the Islands. In the last case we have to weight with the relative fallout and with the production of grain in the respective parts of the country. This probably yields a better estimate than that obtained from the total country models because the production of grain species differs in the different parts of the country. Table 2.2.2.1. shows the various estimates for the entire country. The production weighted models yielded higher sensitivities than the unweighted total country model except in the case of wheat, because approximately 75% of the wheat was grown in the Islands whereas the other species were most frequent in Jutland. However, as the relative standard error of radioecological sensitivities of grain was of the order of 10% (D.2.2.2.2), the differences observed between the various estimates in table 2.2.2.1. are generally immaterial.

On a kg basis the cereal crops were more sensitive to ^{137}Cs than to ^{90}Sr contamination, except for oats which showed a higher sensitivity to ^{90}Sr contamination due to the high root uptake of ^{90}Sr, especially in Jutland.

The prediction models for grain suggested that direct contamination played an important role for the radionuclide content in cereals. SCOTT RUSSELL (Ru66a) has called attention to the importance of so-called floral absorption in connection with the contamination of cereal crops with nuclear debris. Floral absorption is direct contamination during inflorescence. Experiments (VI, VIII) with direct contamination of cereal crops by radiostrontium and radiocesium have shown that cesium

Table 2.2.2.1. Summary of radioecological sensitivities (pCi kg^{-1} y per mCi km^{-2}) in cereal grain from the entire country.

Species	Nuclide	Total country model (tables C.2.2.3. and C.2.2.6.)	Fallout-weighted Jutland and Islands model a)	Fallout- and production-weighted Jutland and Islands model b)
			(tables C.2.2.1. and C.2.2.4.)	
Rye	^{90}Sr	32	27	29
Barley	"	26	25	29
Wheat	"	24	23	20
Oats	"	37	37	43
Rye	^{137}Cs	46	45	48
Barley	"	32	31	34
Wheat	"	28	30	25
Oats	"	27	27	29

a) 1 mCi km^{-2} in Denmark corresponded to 1.113 mCi km^{-2} in Jutland and 0.887 mCi km^{-2} in the Islands.

b) Jutland produced 2/3 of the rye and barley, 1/4 of the wheat and 3/4 of the oats grown in Denmark (Da 57-77).

The calculation of the radioecological sensitivities from the prediction models assumed that 1 mCi ^{90}Sr km^{-2} y^{-1} corresponded to 0.24 mCi ^{90}Sr km^{-2} $(July\text{-}Aug)^{-1}$ and 1 mCi ^{137}Cs km^{-2} y^{-1} ~ 0.54 mCi ^{137}Cs km^{-2} $(May\text{-}Aug)^{-1}$. These relations were the mean values observed since 1962 in Danish fallout.

is translocated to the grain to a far larger extent than radiostrontium, and that for the levels found in the mature grain the time of contamination is thus more important for ^{90}Sr than for ^{137}Cs. Since 1967 a series of experiments (Aa72b, VI VII, VIII) has been performed to investigate the direct contamination of cereal crops with various radionuclides. A study has been made of the translocation of these nuclides and an estimate of the so-called "normalized specific activity", NSA, (Cham70) defined as: (Ci kg^{-1} dry weight of crop) · (Ci d^{-1} m^{-2} of ground)$^{-1}$. The NSA is analogous to the rate factor (cf.C.2.) in the prediction models: (pCi kg^{-1} crop weight) · (mCi km^{-2} $period^{-1}$)$^{-1}$.

When all experimental data collected since 1967 are applied, we obtain the NSA figures shown in table 2.2.2.2. It appears

Table 2.2.2.2. A comparison between experimentally and empirically determined NSA figures for ^{90}Sr and ^{137}Cs in cereal grain. (NSA: (Ci kg^{-1})·(Ci $d^{-1}m^{-2})^{-1}$)

Nuclide	Study	Barley	Wheat
^{90}Sr	exp.	1.3 ± 0.4 (n = 4)	1.3 ± 0.6 (n = 2)
	emp.	2.8 ± 0.2 (n = 5)	3.3 ± 0.6 (n = 5)
^{137}Cs	exp.	5.2 ± 1.0 (n = 5)	7.4 ± 1.3 (n = 2)
	emp.	6.3 ± 0.7 (n = 5)	5.4 ± 0.3 (n = 5)

The error terms are 1 SE estimated from the NSA values from single experiments and from individual state experimental farms for the experimental and empirical values, respectively. The n-values indicate the number of experiments and state experimental farms on which are based the experimental and empirical means, respectively. As the experiments were all performed at Risø, the means of the five state experimental farms for the Islands were used in th. empirical estimates of the NSA values; t-tests between the empirical and the experimental values revealed no significant difference between the two sets except in the case of ^{90}Sr in barley, where the experimental NSA was probably (p = 0.02) lower than the empirical.

The empirical values in the table differed a little from those estimated from tables C.2.2.1. and C.2.2.4., because the former were estimated from individual prediction models for each farm.

that the values for Sr found experimentally were approximately half those determined empirically, MIDDLETON et al. (Mid63) found a similar ratio between experiments and survey data. The experimental NSA figures for Cs were higher for wheat but a little lower for barley than the empirical NSA. Thus the experimentally determined NSA values were generally a little lower than the empirical. This should be recalled if the experimental data are applied for prediction purposes in connection with other nuclides than ^{90}Sr and ^{137}Cs (cf.2.2.3.). However, the experimental NSA values may give the correct answer within a factor of approximately two.

2.2.3. Manganese-54 in cereal grain

During 1963-1965 ^{54}Mn was determined in Danish cereal grain (Aa66c, RRD63, RRD64, RRD65), and it was observed that the contamination of the grain was direct and not, as suggested by SUTTON and KELLY (Su66), indirect. These two authors base

their conclusions on the fact that the ^{54}Mn/stable Mn ratios were nearly constant in different milling fractions of US wheat grown in 1963. However, as has been experimentally shown (VI, VIII), ^{54}Mn deposited directly on the crops is translocated within the plants, and the ^{54}Mn from direct deposition may thus follow the stable manganese distribution in the crops. The anova of ^{54}Mn in cereal grain (table B.2.2.5.) showed significant main effects, but no interactions. The time variation was most pronounced. The ^{54}Mn concentrations in cereal grain thus decreased by a factor of approx. 30 from 1963 to 1965 corresponding to an effective half-life of 0.4 y, which was identical to the effective half-life observed in air samples (1.2.5.). The local variability (table B.2.2.9.) was similar to that of ^{137}Cs (table B.2.2.8.). As regarded the variation among species, rye showed the highest levels (as also observed for ^{137}Cs) but the variability among species was less pronounced for ^{54}Mn; oats thus contained nearly as much ^{54}Mn as rye.

As both the anova and the experiments on direct contamination of grain indicated that ^{137}Cs and ^{54}Mn behaved similarly in cereal grain, the prediction models for ^{54}Mn were assumed to be similar to those for ^{137}Cs, i.e. the concentrations in grain were proportional to the fallout rates in May-August. However, the total deposition from harvest to harvest, i.e. from September to August in the following year, in some cases fitted the data better (table C.2.2.7.). The radioecological sensitivities for ^{54}Mn were on the average 3/4 of those found for ^{137}Cs, which was compatible with a less pronounced translocation of ^{54}Mn as compared with ^{137}Cs (VI, VIII). The experimentally determined NSA values were 2.2 ± 0.8 ($n = 2$) and 2.9 ± 0.2 ($n = 2$) for barley and wheat, respectively. Although these values were lower than those found empirically (6 ± 1 and 5 ± 1 ($n = 5$), they were not significantly different from them (cf.2.2.2.).

The infinite time exposure integral of Danish grain was estimated at 25 pCi ^{54}Mn kg^{-1} y per mCi ^{54}Mn km^{-2}, or (according to (1.2.5.)) as 54 MCi ^{54}Mn resulted in 149 pCi ^{54}Mn m^{-3} y $\sim$ 148 pCi ^{54}Mn l^{-1} y $\sim$ 93 mCi ^{54}Mn km^{-2} (1.3.2.), then 1 MCi ^{54}Mn

released in a megaton test in the northern hemisphere would yield 44 pCi $^{54}Mn\ kg^{-1}$ y in Danish average cereal grain. The corresponding figure for ^{137}Cs was 150 pCi $^{137}Cs\ kg^{-1}$ y per MCi ^{137}Cs, and for ^{90}Sr it was 130 pCi $^{90}Sr\ kg^{-1}$ y per MCi ^{90}Sr.

2.2.4. Strontium-89 in cereal grain

During the high-fallout periods in 1962 and 1963 ^{89}Sr was present in Danish grain samples. An anova revealed no significant variations among species and the data concerning the various species were consequently pooled before the estimation of the NSA. The fallout rates during July-August 1962 and 1963 were 156 and 302 pCi $^{89}Sr\ m^{-2}\ d^{-1}$, respectively (RRD62, RRD63). On this basis, and from the ^{89}Sr concentrations in the grain, the NSA was estimated at 5.5 $\pm$ 1.3 pCi $^{89}Sr\ kg^{-1}$ d per pCi $^{89}Sr\ m^{-2}$. The NSA for ^{89}Sr in grain was thus close to that found for ^{54}Mn and ^{137}Cs, but higher than that for ^{90}Sr, because ^{89}Sr is supposed to be attached to greater particles than is ^{90}Sr (I, II).

Strontium-89 does not play any important role in the contamination of cereal products because the time elapsing between harvest and consumption normally reduces the concentrations of this nuclide to insignificant levels as it has a short half-life (50.5 d). Thus, the relatively high NSA in grain does not manifest itself as a high radioecological sensitivity of the diet to ^{89}Sr contamination.

2.3. Bread

Og den ene henter Hvedebrød og Kringler hos den anden, for fremmed Mad Smager bedst.

HOLGER DANSKE

And they get white bread and biscuits from each other; for other people's food tastes best.

HOLGER THE DANE

In Denmark rye bread is made of rye flour (100% extraction) and white bread of wheat flour (75% extraction). The grain used for the flour does not necessarily come from the area of the country in which the bread is consumed. Furthermore, the flour may not originate from the last harvest. Finally, small

quantities (of the order of 1% (Da57-77)) of the wheat and rye used in the production of bread usually come from abroad. The main purpose of our bread measurements was therefore to elucidate to what extent the cereal grain discussed in the preceding section represents the actual intake of radionuclides from cereal produce.

2.3.1. Variation with time, location and species

The ratios between the radionuclide concentrations (pCi ^{90}Sr kg^{-1} and pCi ^{137}Cs kg^{-1}) in bread collected in June of a given year and the corresponding concentrations in grain from the previous harvest were analyzed by anovas for the period 1962-1973. It was assumed that bread from June was manufactured only from grain grown in the previous year and that all bread was made of locally grown grain. All anovas showed a significant time variation; the ratios were thus in general low when the grain activity levels were relatively high, while high ratios appeared when the grain levels were low. This resulted mainly from the fact that the bread samples had also been prepared from grain older than that of the previous harvest. The local variations were significant too. The ratios from the Islands generally exceeded those from Jutland, because of the transfer of rye from Jutland to make rye bread in the Islands and of wheat from the Islands to make white bread in Jutland (Da57-77). As both ^{90}Sr and ^{137}Cs concentrations were lower in grain from the Islands than from Jutland, the bread/grain activity ratios showed the observed tendency. The anovas finally indicated significant interactions between years and locations, which were among other things ascribed to a changing pattern in the time of the transfer of grain, and thus of activity to the bread, from one part of the country to another. From the beginning of the sixties to the start of the seventies Jutland has thus shown a decreasing relative contribution to the total production of wheat as well as rye in Denmark (Da57-77).

The mean ratios between the activity concentrations in the bread and in the corresponding grain were 0.75 for ^{90}Sr in rye bread, 0.18 for ^{90}Sr in white bread, 0.75 for ^{137}Cs in rye bread and 0.40 for ^{137}Cs in white bread. These ratios were calculated

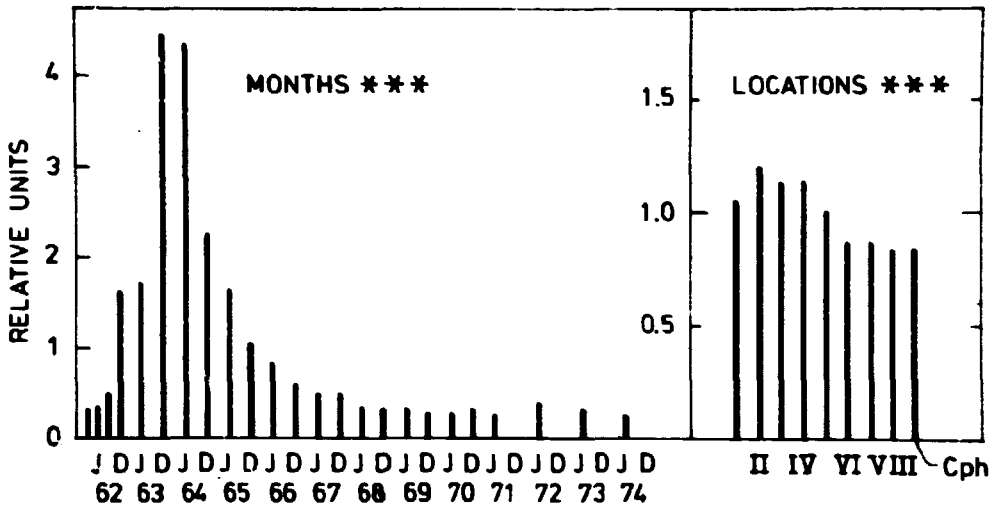

Fig. 2.3.1.1. The variation of pCi ^{90}Sr kg^{-1} in Danish rye bread collected in June and December 1962-1974 (cf. table A.1.2.3.) in the 8 zones and in Copenhagen (cf. figs. A.1.4.2.1. and A.1.4.2.2.). The bars show the concentrations relative to the grand mean 83 pCi kg^{-1} (= 1 at the relative scales).

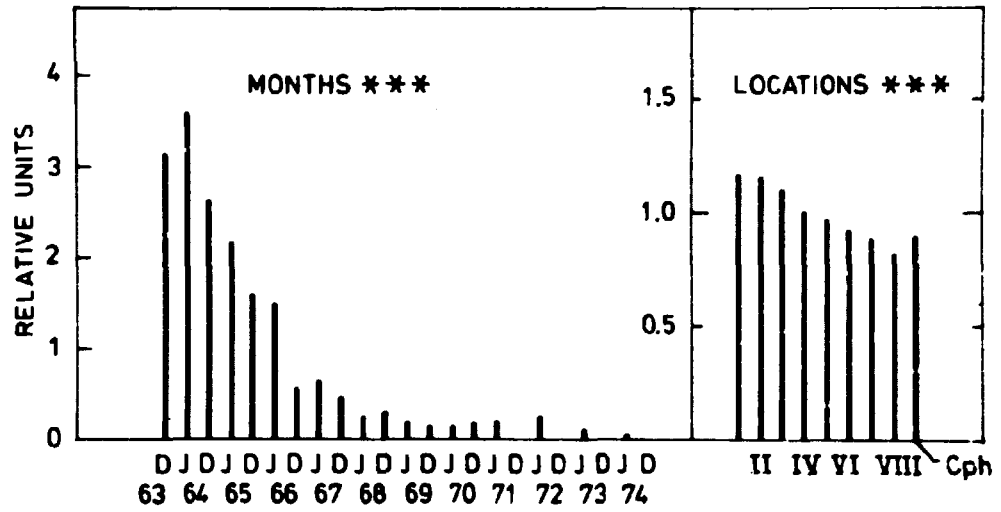

Fig. 2.3.1.2. The variation of pCi ^{137}Cs kg^{-1} in Danish rye bread collected in June and December 1963-1974 (cf. table A.1.2.3.) in the 8 zones and in Copenhagen (cf. figs. A.1.4.2.1. and A.1.4.2.2.). The bars show the concentrations relative to the grand mean 239 pCi kg^{-1} (= 1 at the relative scales).

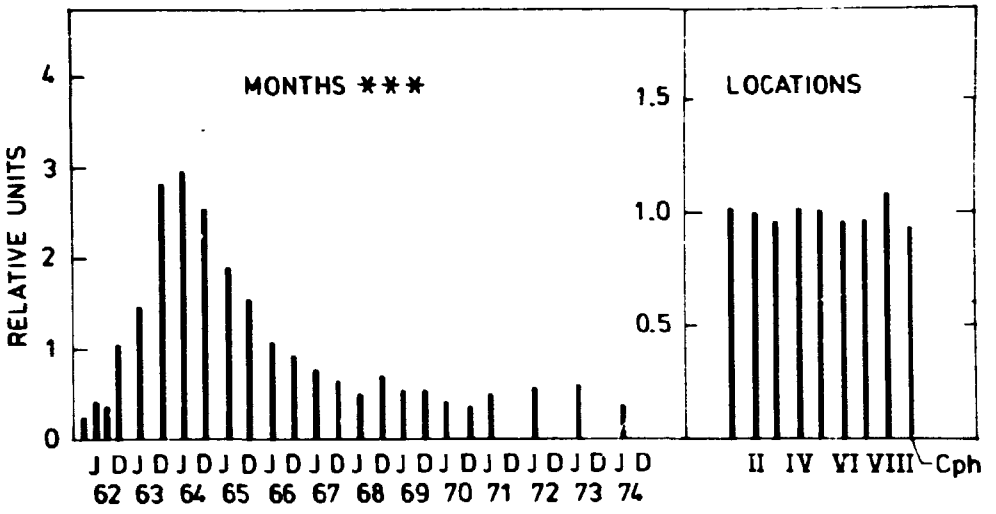

Fig. 2.3.1.3. The variation of pCi ^{90}Sr kg^{-1} in Danish white bread collected in June and December 1962-1974 (cf. table A.1.2.3.) in the 8 zones and in Copenhagen (cf. figs. A.1.4.2.1. and A.1.4.2.2.). The bars show the concentrations relative to the grand mean 13.7 pCi kg^{-1} (= 1 at the relative scales).

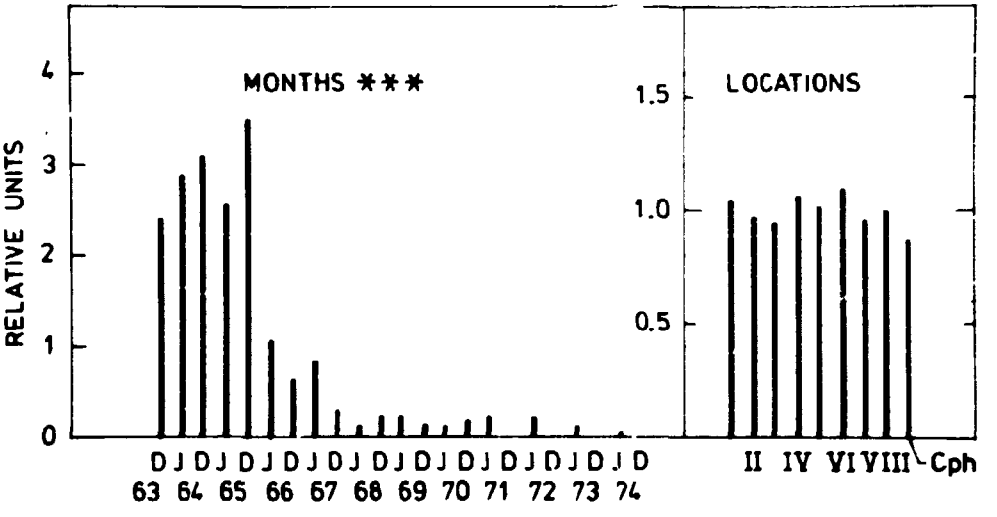

Fig. 2.3.1.4. The variation of pCi ^{137}Cs kg^{-1} in Danish white bread collected in June and December 1963-1974 (cf. table A.1.2.3.) in the 8 zones and in Copenhagen (cf. fig. A.1.4.2.1. and A.1.4.2.2.). The bars show the concentrations relative to the grand mean 75 pCi kg^{-1} (= 1 at the relative scales).

for the period 1962-1973 for ^{90}Sr and for 1964-1973 for ^{137}Cs under the assumption that approx. 2/3 of the rye used for rye bread and 1/4 of the wheat used for white bread were grown in Jutland (Da57-77). The theoretical ratios between bread and grain were 0.74, 0.15, 0.74, and 0.37, respectively. These theoretical ratios were based on the assumption that 1 kg flour yielded 1.35 kg bread (Da68), and that the ^{90}Sr and ^{137}Cs concentrations in flour used for rye bread contained all the activity of the whole grain (100% extraction), while the white bread flour (75% extraction) contained 20% and 50%, respectively, of the concentrations in the whole grain (RRD60, RRD64). The figure of 20% for ^{90}Sr in wheat flour was determined at a time when direct contamination was dominant. Since about one third of the stable Sr is found in white flour (70% extraction) (Un69), it was to be expected that the percentage would increase from 20% towards 33% as the indirect contamination became relatively more important. This would result in an increasing ratio between ^{90}Sr concentrations in bread and in grain from 0.15 to 0.24. Thus we may expect a ratio higher than 0.15 for the entire period, as also observed, and for the long-term situation with decreasing fallout rate we may further expect a ratio approaching 0.24.

An evident result of the transfer of grain from one part of the country to another is the obliterating of the local variation of the activity concentrations of the bread samples (table B.2.3.1. and figs.2.3.1.1.-2.3.1.4.). Although wheat from Jutland thus showed higher ^{90}Sr as well as ^{137}Cs concentrations than wheat from the Islands, the white bread did not show any significant local variation. In the case of rye bread, the local variability of ^{90}Sr and ^{137}Cs was 0.13 and 0.12, respectively, as compared with 0.28 and 0.21 for rye (tables B.2.2.7 and B.2.2.8.).

From December 1963 to December 1965 Danish bread samples were also analyzed for ^{54}Mn (RRD63, RRD64, RRD65). The mean ratio between the ^{54}Mn concentrations in rye bread and rye was 0.77, i.e. in agreement with the theoretical values of 0.74. For wheat and white bread, the mean ratio was 0.09, indicating that the ^{54}Mn concentration of the flour was approx. 10-15% of that

of the whole grain. In their study of milling products of Kansas wheat, SUTTON and KELLY (Su66) found that the ^{54}Mn concentration of wheat flour (extraction rate 0-69%) was 10% of the concentration of the whole grain, i.e. in agreement with the above estimate. We may thus conclude that ^{54}Mn is concentrated in the outer parts of the grain to a greater extent than are both ^{90}Sr and ^{137}Cs. As far as contamination by ^{54}Mn is concerned, the consumption of rye bread (100% extraction) is thus much more important than that of white bread. The consumption of rye bread was in fact the main reason why the Danish diet contained measurable concentrations of ^{54}Mn during 1963-1965.

Since 1964 rye bread and white bread have been collected in June and December in the Faroes. Anovas of the ratios between the radionuclide ^{90}Sr concentrations found in Faroese and in Danish bread did not show any significant variation with time or with bread type. The mean ratios were 0.5 for rye bread and 0.6 for white bread. In the case of ^{137}Cs, Faroese bread also contained less activity than the Danish bread. The mean ratios were 0.6 and 0.9 for rye and white bread, respectively, and the two ratios differed significantly from each other. The lower concentrations in Faroese bread partly resulted from a lower extraction flour used for the rye bread (light rye bread), and partly from the importation of flour from countries other than Denmark, e.g. the U.K. Less than half of the grain imported to the Faroes comes in fact from Denmark (Da70). The foreign flour may in general have contained less activity than the Danish flour, and furthermore the $^{137}Cs/^{90}Sr$ ratio may have been greater in the foreign flour due to root uptake of ^{137}Cs in the foreign cereals. This would explain the relatively high mean ratio found between the ^{137}Cs concentrations in Faroese and Danish white bread as compared to ^{90}Sr. The lower ^{90}Sr and ^{137}Cs concentrations in Faroese bread imply a correction of the estimates of the intakes of the two radionuclides with the total Faroese diet (RPV62-76). For cereal products, these estimates have been based on Danish grain data, which has involved an overestimate of the intake.

Since April 1958 creta praeparata ($CaCO_3$) has been added to

Danish flour. The mean contents were 3.07 ± 0.52 (1 SD) g Ca kg^{-1} rye bread (244 determinations) and 2.07 ± 0.37 g Ca kg^{-1} white bread (236 data) in the period 1960-1975. According to a Government notice of July 29, 1957, (Da68), the contents should be approx. 3 g Ca kg^{-1} rye bread and 1.5 g Ca kg^{-1} white bread. As the natural Ca content (~ 0.3 g Ca kg^{-1} bread) was included in the measurements, the white bread apparently contained a little more of the creta praeparata than recommended, while rye bread on the average contained a little less. There was no time trend in the Ca concentrations of the bread, but it appeared that the Ca content of white bread from Lolland-Falster was 25% higher than in white bread from West Jutland. Milk is often used in the manufacture of bread (Da68), and this may explain the observed variations of the calcium levels.

Faroese bread contained approx. half as much Ca as Danish, viz. 1.72 ± 0.73 (1 SD) g Ca kg^{-1} rye bread (25 data) and 0.99 ± 0.48 g Ca kg^{-1} white bread (25 data) during 1964-1975. In some cases the pCi ^{90}Sr $(g\ Ca)^{-1}$ levels may thus be higher in Faroese bread than in Danish, although the pCi ^{90}Sr kg^{-1} levels were generally lower in Faroese bread.

2.3.2. Prediction models and relations

As the radionuclide concentrations in bread were not strictly proportional to the concentrations in grain, prediction models were obviously calculated from the bread data. The transfer factors from grain to bread were estimated from tables C.2.3.1. and 2.2.2.1. For ^{90}Sr in rye bread, the factor was 0.79, for ^{137}Cs: 0.79; in the case of white bread, 0.21 of the ^{90}Sr and 0.40 of the ^{137}Cs were transferred from the wheat to the bread. These estimates were in agreement with the theoretical values mentioned above. It appeared that Danish bread was more sensitive to environmental contamination with ^{137}Cs than with ^{90}Sr, which is partly a result of the greater sensitivity of grain to ^{137}Cs contamination (table 2.2.2.1.), and partly of the greater translocation of ^{137}Cs within the grain.

Although the Faroese bread was not made solely from Danish grain, its concentrations may be related to the Danish fallout

data as shown in table C.2.3.1. The ratios between the radioecological sensitivities of Faroese and Danish bread were 0.62 in the case of rye bread and 0.86 for white bread (for both ^{90}Sr and ^{137}Cs). The ratio for ^{90}Sr between Faroese and Danish white bread was higher than found by the measurements made hitherto (2.3.1.) because the soil factor in the models carried a relatively heavy weight when the infinite time integrals were calculated. As the soil factors for Danish and Faroese white bread were nearly identical, the Faroese ^{90}Sr bread levels would have to approach the Danish.

2.3.3. Other grain products

Oat grits have been collected regularly through the years and analyzed for ^{90}Sr and ^{137}Cs. From the prediction models for grits (table C.2.3.1. Nos. 10 and 11) and from table 2.2.2.1. (last column), the transfer factor from oats to grits was estimated at 0.33 for ^{90}Sr and 0.45 for ^{137}Cs. In the calculations of the ^{90}Sr and ^{137}Cs concentrations in grits from oats (RRD59-76), the transfer factors 0.4 and 0.75 for ^{90}Sr and ^{137}Cs, respectively, have been used hitherto. Hence, the estimates of ^{137}Cs in grits have been 67% too high and estimates of ^{90}Sr 20% too high. The calcium content in grits, which also contained creta praeparata, was measured at 3.3 ± 0.2 g Ca kg^{-1} (1 SD) (RRD59-76).

Rice was also analyzed for ^{90}Sr and ^{137}Cs, but the data were too few and scattered to calculate any prediction models. The concentrations of ^{90}Sr and ^{137}Cs in polished rice were in general very low. The infinite time integrated levels for the period up to 1975 were estimated to be of the order of 1 pCi kg^{-1} y per mCi km^{-2} for ^{90}Sr as well as for ^{137}Cs. As there was no information on deposition from the area where the rice was grown, the Danish data for deposition of 73 mCi ^{90}Sr km^{-2} during 1950-1975 was used in the estimate.

2.4. Grass and other fodder crops

Hun flettede sig en Seng af Straastraa og hang den under et stort Skrappeblad, så kunde det ikke regne på hende;
TOMMELISE

She took blades of grass and plaited herself a bed, which she hung under a large dock-leaf, so as to be out of the rain.
THUMBELINA

2.4.1. Grass

Grass is a useful collector of direct radioactive contamination because its large surface-to-volume ratio retains nuclear debris with good efficiency, and because it is generally easy to obtain. Furthermore, because it is an important constituent of cattle fodder, and thus influences the radioactivity concentrations encountered in cows' milk and in beef, grass was included right from the start in the fallout measurements carried out at Risø (He56-57). The first UNSCEAR report (Un58) presented some of the earliest results of ^{90}Sr and ^{137}Cs determinations on grass and hay collected in 1954-1957 in the U.S. and the U.K.

The samples in the present study were collected throughout the year at Risø and during the grazing period at the state experimental farms. In the Faroes and Greenland, grass was obtained during the summer (cf.fig.2.4.1.). The higher ^{90}Sr concentrations in Faroese grass were partly explained by the higher fallout levels in the Faroes, partly by the higher direct contamination of the Faroese pastures because of their lower productivity (Cham70), and partly by the higher indirect contamination, which is due among other things to the lower Ca content (Mi60) of Faroese soil. The last two reasons for the enhanced levels also apply to grass samples from Greenland. The variability among years (table B.2.4.1.) was generally higher for Denmark than for the Faroes and Greenland, because direct contamination of grass was more important in Denmark. In the case of ^{137}Cs, the indirect contamination on the soils rich in organic matter (Fr63, Ba64) played an important role for the Faroese and Greenlandic grass samples, whereas the root uptake of ^{137}Cs from the mineral-rich Danish soils was a secondary factor. The local variability of ^{90}Sr in Danish grass was similar to that observed for precipitation (0.22 against 0.18) suggesting the indirect contamination of grass with ^{90}Sr to be proportional to the direct contamination and thus rela-

tively unaffected by the type of soil; this was in the period 1962-1970, in 1977 (RKD77), however, the local variability was 0.32 showing the increasing importance of root uptake. ^{137}Cs, however, showed a higher variability among locations than ^{90}Sr; this may be due to indirect ^{137}Cs contamination of grass at some locations, while others rely on direct contamination only.

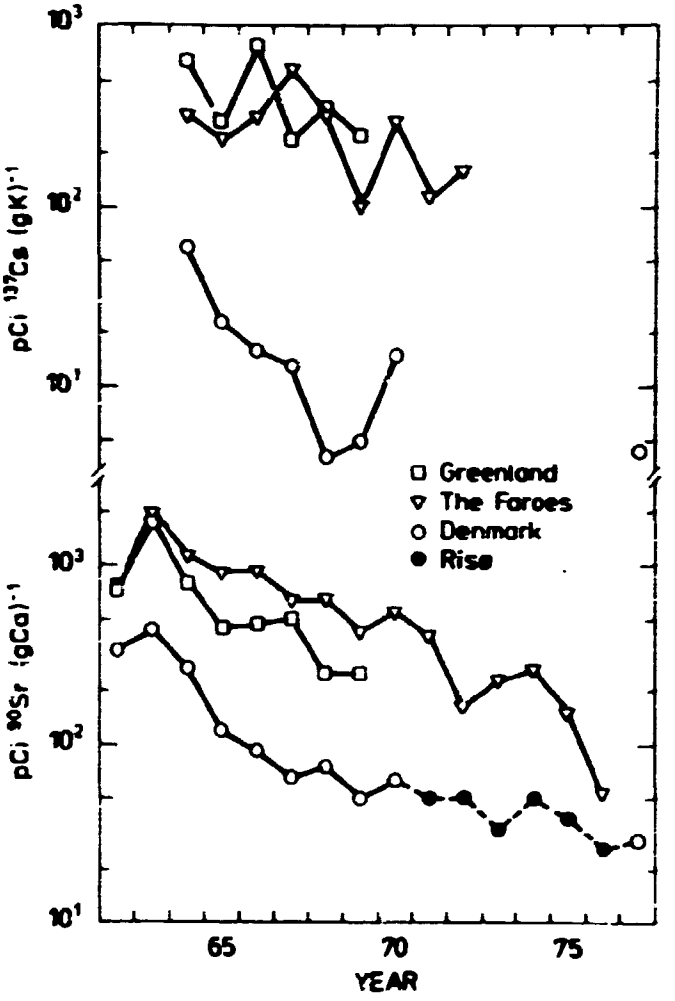

Fig. 2.4.1. The medians of the ^{90}Sr and ^{137}Cs levels in Danish, Faroese and Greenlandic "summer grass" collected May-October since 1962 (cf. table A.1.2.4.).

An anova of all ^{90}Sr data on Danish grass showed a significant variation with time and location. Throughout the years, the highest ^{90}Sr concentrations normally occurred in the first quarter of the year just before the maximum fallout levels normally appeared. This apparent pecularity was due to the rapid increase in growth rate of the grass in the second quarter of the year. The sudden increase in the productivity of the pastures was tantamount to a reduced susceptibility to

direct contamination (Cham70) and an enhanced field loss*) of deposited activity (Chad70). Although the fallout rate thus increased, the net result was decreasing grass concentrations of fallout nuclides.

The radioecological sensitivity of summer grass (table C.2.4.1.) was 31 pCi ^{90}Sr (g Ca)$^{-1}$ y (or 38 pCi ^{90}Sr kg^{-1} y) per mCi ^{90}Sr km^{-2} (D.2.4.1.) in Denmark. The corresponding values for the Faroes and Greenland were 216 (or 268) and 442 (or 548), respectively. The Faroese and Greenlandic grass was thus nearly 7 and 14 times more sensitive to contamination with ^{90}Sr than Danish grass. In the case of ^{137}Cs, the radioecological sensitivity (table C.2.4.2.) of Danish grass was 5.1 pCi ^{137}Cs (g K)$^{-1}$ y (or 24 pCi ^{137}Cs kg^{-1} y) per mCi ^{137}Cs km^{-2}. In the grass from the Faroes and Greenland, the corresponding values were 76 (or 524) and 79 (or 352), respectively. The radioecological sensitivities estimated for Greenlandic grass were approximate as the data were scarce; especially the estimates of the pCi kg^{-1} sensitivities were uncertain, because the Ca and K concentrations in the Greenlandic (and Faroese) samples showed great variations (cf.D.2.4.1.). The radioecological sensitivities of Faroese and Greenlandic grass may thus be nearly similar (cf. also the observations for sheep, 3.4.3.).

The normalized specific activity (NSA) was calculated for the Danish grass samples: 45 pCi ^{90}Sr kg^{-1} (dry matter) per pCi ^{90}Sr m^{-2} d^{-1} and 20 pCi ^{137}Cs kg^{-1} (dry matter) per pCi ^{137}Cs m^{-2} d^{-1}. CHAMBERLAIN (Cham70) and ERIKSSON (Er77) quoted similar values for summer grass. CHAMBERLAIN found that winter grass showed 2-3 times higher NSA values, which is also in agreement with the observations in table C.2.4.1. (Nos.1, 2 and 3) where the ratio between the radioecological sensitivities of winter grass and summer grass was 2.3.

2.4.2. Fodder crops other than grass

Besides grass, the main constituents of Danish cattle fodder

*) Field loss is the loss of activity from crops due to reasons other than radioactive decay.

Table 2.4.2. Transfer factors for ^{90}Sr and ^{137}Cs in cattle feed. (Fodder class 5 ~ 11 kg milk d^{-1} (4.25% fat) 9 feed units)*). (Br 69).

		Grass and hay (as fresh)	Roots 11% dry matter	Leaves of roots (silage)	Straw	Barley (concentrates)	Total intakes pCi ^{90}Sr per cow per mCi ^{90}Sr km^{-2}	Total intakes pCi ^{137}Cs per cow per mCi ^{137}Cs km^{-2}
pCi kg^{-1} days per mCi km^{-2}	^{90}Sr	14030	4944	40515	95083	9508		
	^{137}Cs	8760	292	4198	22995	11498		
Winter plan No 18	kg d^{-1} per cow (feed units)	5.5 (1)	45 (4.5)	12 (1)	2 (0.5)	2 (2)	$1.00 \cdot 10^6$	$1.81 \cdot 10^5$
Winter plan No 1	kg d^{-1} per cow (feed units)	- -	25 (2.5)	60 (6.5)	1 (~0)	- -	$2.65 \cdot 10^6$	$2.82 \cdot 10^5$
Winter plan No 46	kg d^{-1} per cow (feed units)	33 (6)	20 (2)	- -	- -	1 (1)	$0.55 \cdot 10^6$	$3.06 \cdot 10^5$
Winter plan No 31	kg d^{-1} per cow (feed units)	- -	57 (5.5)	- -	4 (1)	2.4 (2.4)	$0.68 \cdot 10^6$	$1.36 \cdot 10^5$
Summer plan No 8	kg d^{-1} per cow (feed units)	44 (8)	- -	- -	- -	1 (1)	$0.63 \cdot 10^6$	$1.97 \cdot 10^5$

*) The milk production corresponded to that in the middle of the period 1962-1974. At present (1979) Danish cows produce approx. 13 kg milk d^{-1} and the fodder class is correspondingly higher.

are roots, root leaves (silage), straw and concentrates (mostly barley grain) (Br69). The sampling of roots, leaves and straw has not been systematic, and prediction models for these items were consequently partly based on related samples. Tables C.2.4.3. and C.2.4.4. show the models estimated.

Based on the fodder mixtures recommended for Danish cows (Br69), the infinite time-integrated intakes of ^{90}Sr and ^{137}Cs with cattle feed were estimated as shown in table 2.4.2. The intakes were calculated for an average milk yield of 11 kg y^{-1} (4.25% fat) (Da57-77). It appeared that the highest ^{90}Sr intakes originate from fodder rich in silage and the lowest from grass diets. As regarded ^{137}Cs, grass diets yield a two to three times higher intake than root diets. The $^{137}Cs/^{90}Sr$ ratio of the grass and hay diet was higher than in the root and silage diet. These variations influenced the local and seasonal variations of ^{90}Sr and ^{137}Cs concentrations in Danish milk (cf.3.2.1.).

2.5. Vegetables and fruit

Saa kunde de see, at det var en rig-tig Prindsesse, da hun gjennem de tyve Madrasser og de tyve Edderdunns Dyner havde mærket Ærten.
PRINDSESSEN PAA ÆRTEN

So then of course they could see that she really was a Princess, because she had felt the pea right through the twenty mattresses and the twenty feather-beds.
THE PRINCESS AND THE PEA

After cereals and milk products, vegetables and fruit have been the most important ^{90}Sr donors in the Danish diet. This group contributed 17 $\pm$ 6% (1 SD) to the total ^{90}Sr intake; with respect to ^{137}Cs, the contribution was 10 $\pm$ 3%. During periods with a high fallout rate, such as 1962-1964, the relative contributions of ^{90}Sr and ^{137}Cs from vegetables and fruit were lower than when the accumulated fallout was high compared to the fallout rate (X).

The first samples of vegetables for ^{90}Sr and ^{137}Cs analysis were collected in 1956 (Un58) in the United Kingdom, the United States and in Japan. Danish investigations of radioactive contamination in vegetables began in 1959. Since then potatoes, white cabbage, carrots and apples have been collected regularly

throughout the years from all parts of the country. In 1960, 1962 and 1966 nearly all species of Danish fruit and vegetables were sampled.

2.5.1. Variations with time, location and species

The time variation for ^{90}Sr in vegetables has in general been less pronounced than in grain and grass. Potatoes, cabbage and carrots have thus all shown relatively slowly decreasing ^{90}Sr concentrations since the maximum years of 1963-1964; by 1975 the levels in these products were approx. two-thirds of the maximum (fig.2.5.1.1.). The variability among years (table B.2.5.3.) was approx. 0.2 (1962-1974) or similar to that observed for ^{90}Sr in soil.

Compared to this, ^{90}Sr in grain and in grass showed variabilities of 0.8 and 1.0, respectively, in the same period. This clearly demonstrates the relatively strong dependence of vegetables on the accumulated ^{90}Sr in the soil. The surfaces of grass and grain exposed to direct contamination are much greater per weight unit than that of the vegetables mentioned above and this enhances direct contamination and with it the time variability of grain and grass. In the case of potatoes and carrots, the edible parts are furthermore hidden below the soil surface, which thus provides an additional protection against direct contamination. Vegetables with a relatively large above-soil surface, such as kale, depend to a greater degree on direct contamination than, e.g., cabbage. From 1963 to 1976 the ^{90}Sr concentration in kale thus decreased by a factor of four.

In apples, the ^{90}Sr concentration was closely related to the fallout rate. From 1963 to the first half of the seventies the levels thus decreased by a factor of approx. 15 ($CV_{p\ years}$ = 0.99).

The ^{137}Cs concentrations in vegetables and fruits varied primarily with the fallout rate. The concentrations in cabbage, potatoes and carrots all decreased by an order of magnitude from 1963 to the first half of the seventies (fig.2.5.1.2.). The variability among years was 1.0-1.6 for these products

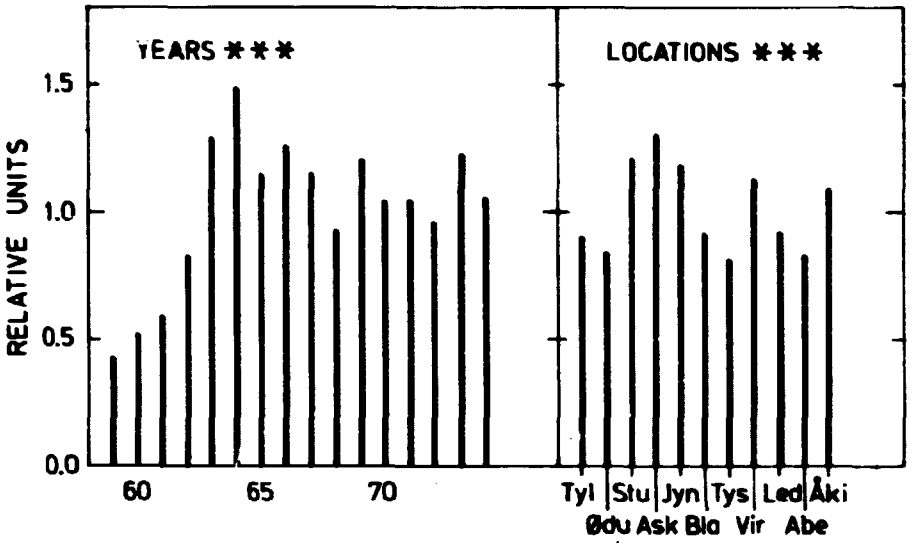

Fig. 2.5.1.1. The variation of pCi ^{90}Sr kg^{-1} in potatoes collected in 1959-1974 at the state experimental farms (fig. A.1.1.3.1.). The bars indicate the concentrations relative to the grand mean 3.1 pCi kg^{-1} (= 1 at the relative scale).

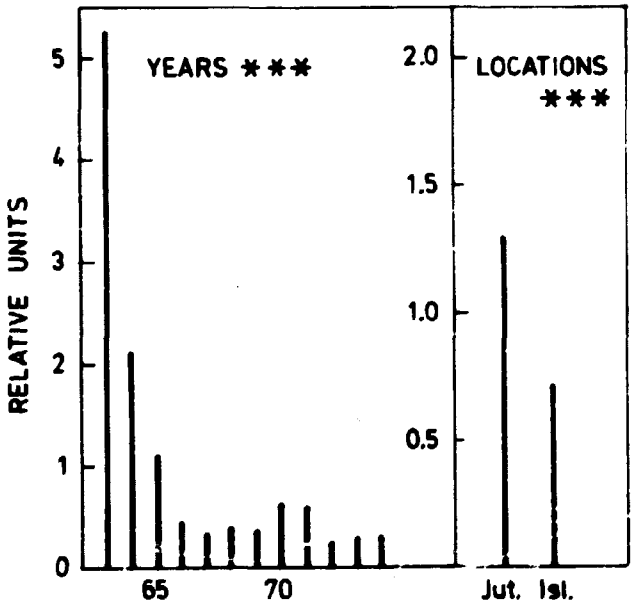

Fig. 2.5.1.2. The variation of pCi ^{137}Cs kg^{-1} in potatoes collected in 1963-1974 at the state experimental farms in Jutland and the Islands (fig. A.1.1.3.1.). The bars show the concentrations relative to the grand mean 19 pCi kg^{-1} (= 1 at the relative scale).

(table B.2.5.2.), i.e., significantly higher than for ^{90}Sr. Apples showed an annual variability of 1.32 during 1963-1974.

In a special study carried out in 1962 and 1963 (RRD62, RRD63) the large basal leaves of brussels sprouts plants were collected and analyzed for ^{90}Sr, ^{137}Cs and stable Sr during the

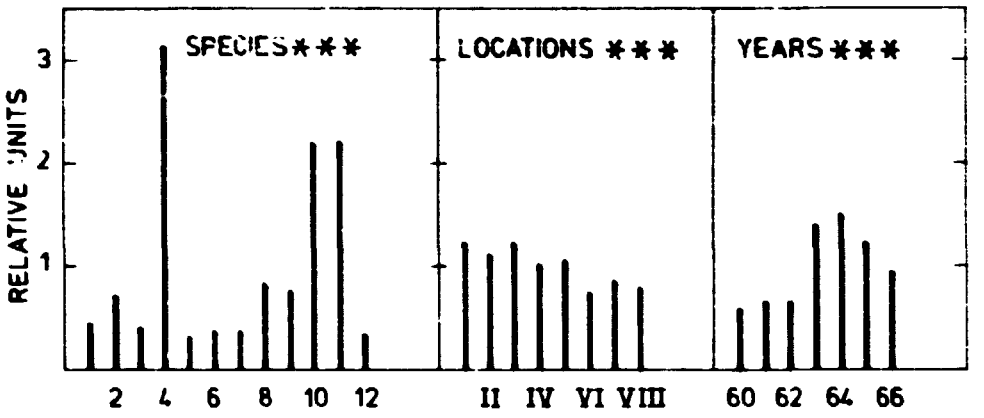

Fig. 2.5.1.3. The variation of pCi $^{90}Sr\ kg^{-1}$ in leaf vegetables collected in 1960-1966 in the 8 zones (cf. table A.1.2.5.2.). The bars show the concentrations relative to the grand mean 23 pCi kg^{-1} (= 1 at the relative scale). The species were: 1: white cabbage, 2: spring cabbage, 3: red cabbage, 4: kale, 5: cauliflower, 6: cucumber, 7: peas, 8: beans, 9: lettuce, 10: spinach, 11: parsley, 12: brussels sprouts.

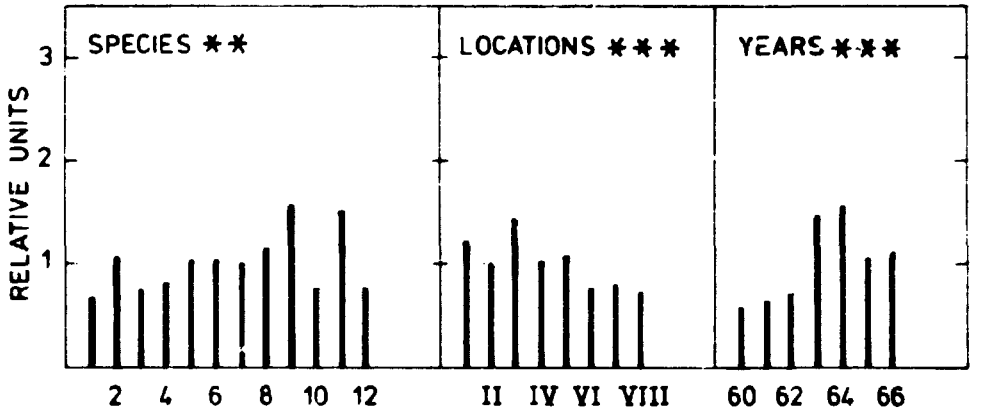

Fig. 2.5.1.4. The variation of pCi $^{90}Sr\ (g\ Ca)^{-1}$ in leaf vegetables collected in 1960-1966 in the 8 zones (cf. table A.1.2.5.2.). The bars show the concentrations relative to the grand mean 30 pCi $(g\ Ca)^{-1}$ (= 1 at the relative scale). For identification of the species, cf. caption to fig. 2.5.1.3.

growing season from six locations distributed throughout Denmark (fig.2.5.1.11.). The study showed that within the growing season, i.e. from June to December, there was only a probably significant difference between the monthly ^{90}Sr levels ($CV_{p\ month} = 0.10$). The concentrations in December were thus probably higher than those of the other months. However, the stable Sr to calcium ratio in the leaves showed a highly significant seasonal variation ($CV_{p\ month} = 0.25$), and so did

the ^{137}Cs concentrations ($CV_{p\ month}$ = 0.24). The highest concentrations of stable Sr were found in mid summer, the lowest (approx. half the summer levels) in winter. The $^{89}Sr/^{90}Sr$ ratio in the leaves indicated that about 1/4 of the ^{90}Sr came from direct contamination in 1962-1963 and 3/4 from root uptake from the soil. The studies of grass (2.4) show that winter grass is more sensitive to direct contamination than summer grass. If the same applies to the leaves of brussels sprouts, we may expect relatively higher direct contamination during the winter than in the summer, as also observed for ^{137}Cs and to some degree for ^{90}Sr. If, however, the Sr/Ca quotient from indirect contamination is lower during the winter, as indicated by the stable Sr/Ca quotient, the winter ^{90}Sr levels may be observed

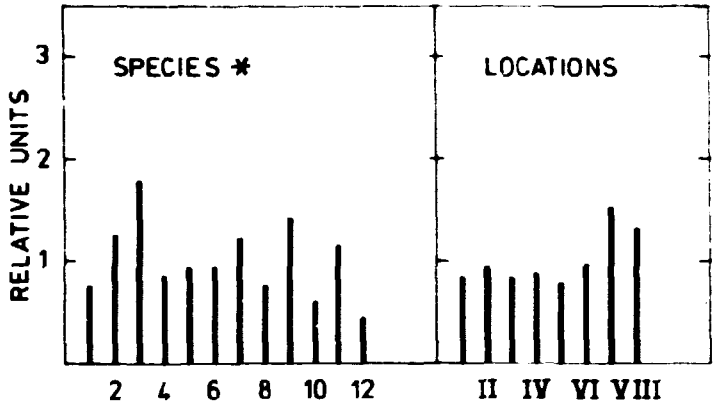

Fig. 2.5.1.5. The variation of stable Sr in leaf vegetables shown in figs. 2.5.1.3. and 2.5.1.4. The bars show the concentrations relative to the grand mean 2.9 mg (g Ca)$^{-1}$ (= 1 at the relative scale). For identification of the species see caption to fig. 2.5.1.3.

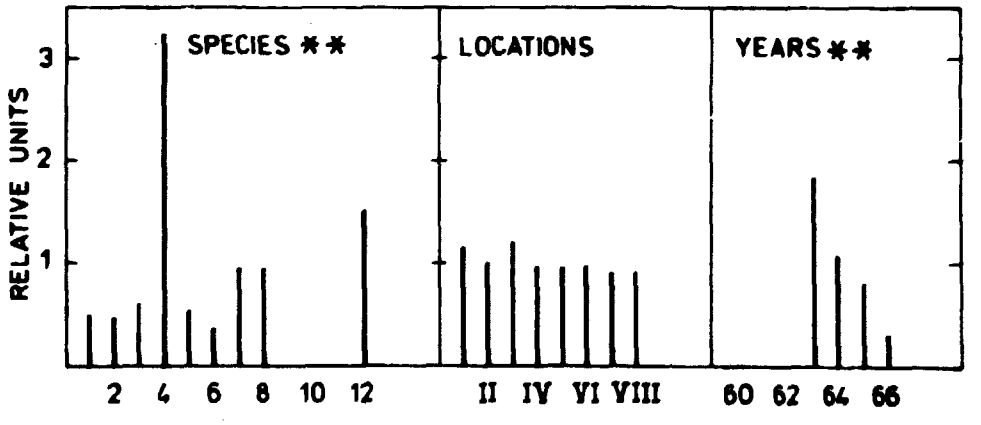

Fig. 2.5.1.6. The variation of pCi ^{137}Cs kg^{-1} in leaf vegetables collected in 1963-1966 in the 8 zones (cf. table A.1.2.5.2.). The bars show the concentrations relative to the grand mean 27 pCi kg^{-1} (= 1 at the relative scale). For identification of the species, cf. caption to fig. 2.5.1.3.

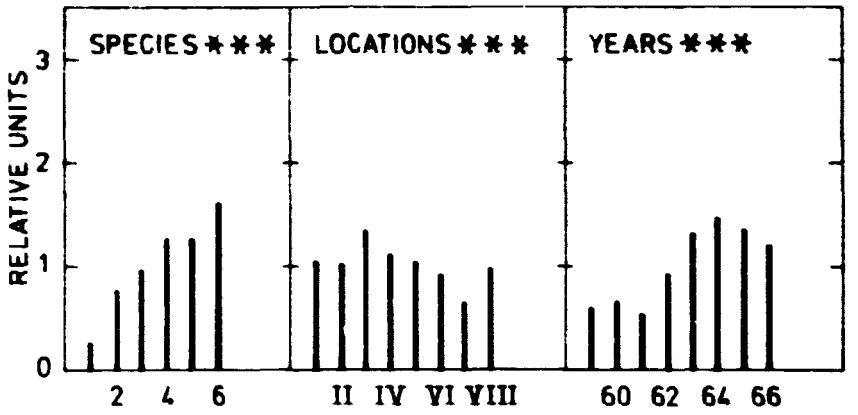

Fig. 2.5.1.7. The variation of pCi ^{90}Sr kg^{-1} in root vegetables collected in 1959-1966 in the 8 zones (cf. table A.1.2.5.2.). The bars show the concentrations relative to the grand mean 14.4 pCi kg^{-1} (= 1 at the relative scale). The species were: 1: potatoes, 2: carrots, 3: onion, 4: leek, 5: beetroot, 6: celeriac.

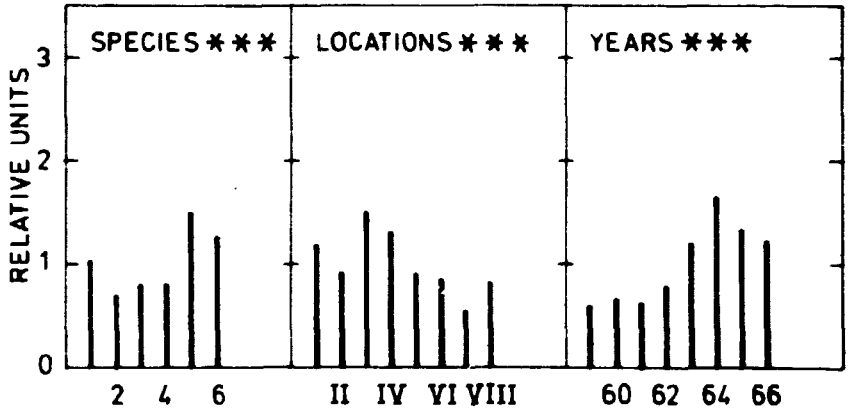

Fig. 2.5.1.8. The variation of pCi ^{90}Sr $(g\ Ca)^{-1}$ in root vegetables collected in 1959-1966 in the 8 zones (cf. table A.1.2.5.2.). The bars show the concentrations relative to the grand mean 54 pCi $(g\ Ca)^{-1}$ (= 1 at the relative scale). For identification of the species, cf. caption to fig. 2.5.1.7.

to increase less rapidly during the winter months than expected solely from the direct contamination.

As regards the local variation in vegetables and fruit, Jutland showed generally higher ^{90}Sr concentrations than the Islands (figs.2.5.1.3, 2.5.1.4, 2.5.1.7 and 2.5.1.8.). The local variability was 0.17 for cabbage and potatoes, and 0.33 for carrots. Grass showed a local variability of 0.22 and grain of $\sim$ 0.4. In the study of brussels sprouts the local variability of ^{90}Sr was 0.31, the two stations in Jutland showing 1.8

times the mean concentration in the Islands. The difference in soil characteristics between the stations in Jutland and in the Islands enhanced the difference between the local ^{90}Sr concentrations in the leaves of brussels sprouts. As was the case for the time variation of ^{90}Sr in leaves of brussel sprouts, the local variation may also have been reduced because the stable Sr to Ca ratios in the leaves were to some degree inversely related to the fallout rate, i.e. the highest mg Sr/g Ca quotients were found in the Islands (fig.2.5.1.12; cf. also figs.2.5.1.5 and 2.5.1.9.). As regards the local variation of the ^{137}Cs concentrations in root vegetables (fig.2.5.1.10.), Jutland showed higher levels than those of the Islands ($CV_{p\ locations} = 0.24$), while leaf vegetables including peas,

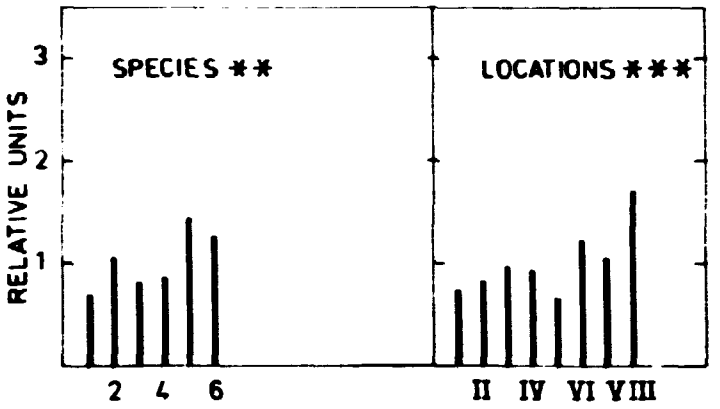

Fig. 2.5.1.9. The variation of stable Sr in root vegetables shown in figs. 2.5.1.7. and 2.5.1.8. The bars show the concentrations relative to the grand mean 4.6 mg (g Ca)$^{-1}$ (= 1 at the relative scales).

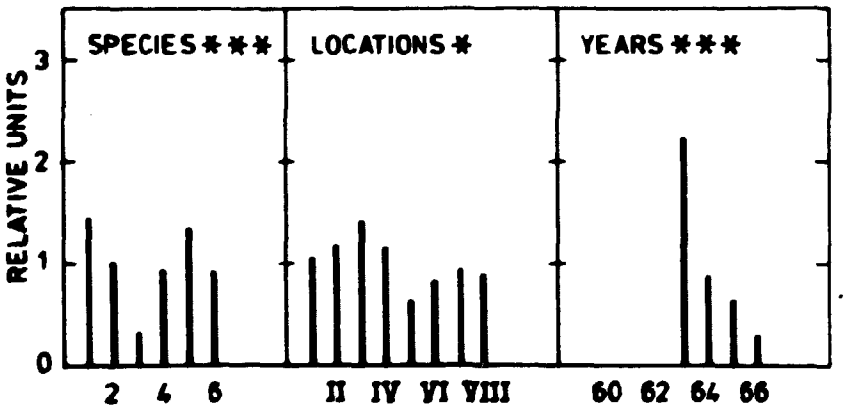

Fig. 2.5.1.10. The variation of pCi ^{137}Cs kg^{-1} in root vegetables collected in 1963-1966 in the 8 zones (cf. table A.1.2.5.2.). The bars show the concentrations relative to the grand mean 26 pCi kg^{-1} (= 1 at the relative scale). For identification of the species, cf. caption to fig. 2.5.1.7.

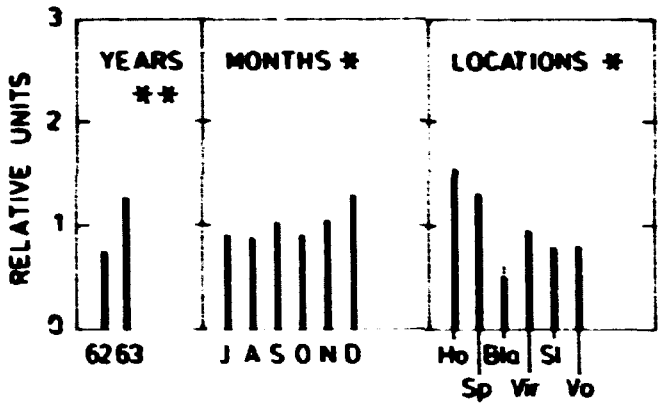

Fig. 2.5.1.11. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in leaves of brussels sprout plants collected monthly (July-December) (cf. table A.1.2.5.2.) at 6 experimental farms (cf. fig. A.1.2.5.2.) during 1962-1963. The bars show the levels relative to the grand mean 37 pCi (g Ca)$^{-1}$ (= 1 at the relative scale).

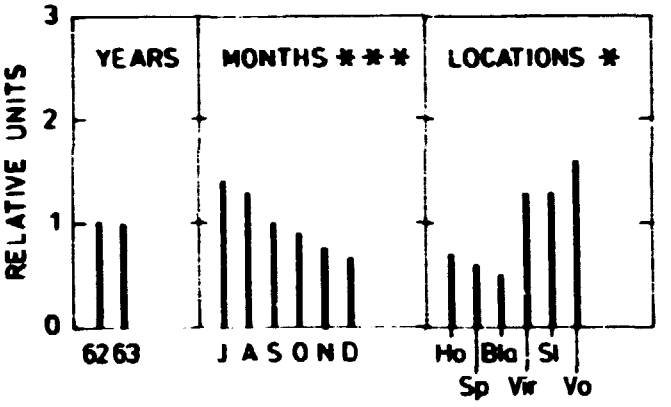

Fig. 2.5.1.12. The variation of stable Sr in the brussels sprout leaves shown in fig. 2.5.1.11. The bars show the levels relative to the grand mean 2.4 mg (g Ca)$^{-1}$ (= 1 at the relative scale).

beans and cucumbers showed a local variability of only 0.11 (fig.2.5.1.6.). This suggested some root uptake of ^{137}Cs from the soils in Jutland. ANDERSEN (An67a) found that root crops tended to accumulate more ^{137}Cs than other crops; root crops may thus show a greater local variability than leaf vegetables. Anovas of the ^{90}Sr and ^{137}Cs levels in fruit (apples and soft fruit) showed no significant local variations, although the concentrations found in Jutland generally seemed to be a little higher than those in the Islands.

The interspecific variations of radionuclide concentrations depend upon the morphology and the physiology of the plants. Morphology plays an important role in direct contamination and

physiology in indirect contamination. ANDERSEN (An67a) studied the indirect contamination of Danish vegetables; he found that the $^{89}Sr/Ca$ ratio decreased from root to top, because plants were able to discriminate against Sr relative to Ca during the translocation of these elements within the plants; the $^{89}Sr/Ca$ ratios in leaves were approx. two-thirds of those in roots. In agreement herewith the present study showed that the stable Sr to Ca mean quotient in green leaf vegetables was approx. two-thirds of the quotient found in root vegetables.

ANDERSEN furthermore observed that the root uptake of Sr by different plant species was related to Ca uptake, i.e., a high Ca uptake was tantamount to a relatively high Sr root uptake. As regards the ^{90}Sr concentrations (pCi kg^{-1} fresh weight) in leaf vegetables, kale and spinach showed the highest levels due to their high Ca concentrations (figs.2.5.1.3. and 2.5.1.4.). The calcium levels in root vegetables and fruit were generally lower, consequently the ^{90}Sr content per kg of vegetables and fruit was lower too. The pCi ^{90}Sr $(g\ Ca)^{-1}$ ratios showed less variation among different species as well as within the same species than the pCi kg^{-1} figures. The variability among root vegetables (table B.2.5.3.) was thus 0.89 for pCi ^{90}Sr kg^{-1} but 0.23 for pCi ^{90}Sr $(g\ Ca)^{-1}$ only, and for leaf vegetables the corresponding values were 0.88 and 0.24, respectively. The higher variation of the pCi kg^{-1} figures within a single species resulted mainly from the varying water content of the samples. However, this variation also occurs when the products are used for consumption, and it is thus not pertinent to relate the activity levels to the dry matter content. It is notable that the ^{90}Sr levels in vegetables were more dependent upon the Ca content of the crops than was the case for grain (cf.figs. 2.2.1.1. and 2.2.1.2.), because root uptake of ^{90}Sr is more important for vegetables than for grain.

The ^{137}Cs concentrations also varied among species, but the variability among species was less than the corresponding variability for ^{90}Sr (cf.table B.2.5.3.) because ^{90}Sr was influenced not only by direct contamination, as ^{137}Cs was primarily, but also by indirect contamination and this enhanced

the interspecific differences for ^{90}Sr. The variability among species was nearly twice as high for the pCi ^{137}Cs kg^{-1} figures than for the ^{137}Cs (g K)$^{-1}$ ratios. Analogous to ^{90}Sr and calcium, the ^{137}Cs concentrations in vegetables were thus influenced by the congener, potassium, in such a way that high concentrations of the congener were tantamount to enhanced concentrations of the radionuclide. With respect to tree fruit such as apples, pears and plums, the ^{137}Cs levels were significantly greater than the ^{90}Sr concentration (pCi kg^{-1} figures). This was contrary to the observations for most vegetables and soft fruit. Tree fruit were primarily contaminated by direct contamination, but whereas ^{137}Cs was translocated from other parts of the tree to the fruit, this was not the case for ^{90}Sr. Consequently, the ^{137}Cs concentration in apples became greater than the ^{90}Sr concentration, because the "effective area" exposed to direct contamination was greatest for ^{137}Cs.

The anovas showed significant interactions between years and locations for leaf vegetables both for ^{90}Sr and for ^{137}Cs. For root vegetables, this interaction was significant for ^{90}Sr only. The interaction between species and locations was significant for ^{90}Sr in root vegetables only. Interactions were to be expected due to the varying contributions from direct and indirect contamination (cf. grain, table 2.2.1.).

2.5.2. Prediction models

For cabbage, carrots, potatoes and apples, the prediction models were calculated for ^{90}Sr and ^{137}Cs both per kg and per g of the congeners Ca and K, respectively (tables C.2.5.1. - C.2.5.4.). With respect to ^{90}Sr, Jutland did not show higher radioecological sensitivity than the Islands except in the case of carrots; however, the correlation between observed and calculated values was poor for carrots from the Islands. Potatoes from Jutland were probably more sensitive to ^{137}Cs contamination than potatoes from the Islands, which suggests a slight "root" uptake from the soils in Jutland.

Vegetables were in general more sensitive to contamination with ^{90}Sr than with ^{137}Cs (pCi kg^{-1}); cabbage and carrots thus

showed a 5-6 times higher sensitivity to the former nuclide. Potatoes, however, were approx. 1.5 times more sensitive to ^{137}Cs than to ^{90}Sr, while apples showed a radioecological sensitivity to ^{137}Cs that was approx. 3 times higher than that observed for ^{90}Sr.

Table C.2.5.5. gives prediction models for some less frequently sampled vegetables and fruit. Kale was the species most sensitive to radioactive contamination with ^{90}Sr as well as with ^{137}Cs - primarily because of its large surface and the subsequent susceptibility to direct contamination. As compared with grain and grass, vegetables and fruit were in general less sensitive to contamination. The transfer factors for ^{137}Cs in grain and grass were thus an order of magnitude higher than those for vegetables, and the ^{90}Sr factors were 2-3 times higher.

It appeared that the correlations between observed and calculated pCi kg^{-1} figures generally were superior to those of the pCi ^{90}Sr (g Ca)$^{-1}$ and pCi ^{137}Cs (g K)$^{-1}$ quotients. This suggested that possible variations in the water content of the samples were less important than the analytical errors involved in the Ca and K determinations.

2.5.3. Iodine in green leaf vegetables

In the western diet vegetables and fruit have been of minor importance as a source of ^{131}I as compared with fresh milk (Ga66). Measurements have therefore been few. After a Chinese atmospheric test explosion in October 1976 (RRD76), a sample of cabbage, one of brussels sprouts and five of kale were collected in Zealand and measured for ^{131}I, as both fresh and cooked samples. Cabbage contained no ^{131}I, brussels sprouts contained 31 pCi ^{131}I kg^{-1} and kale 127 pCi ^{131}I kg^{-1} (median 84 pCi kg^{-1}). The cooked kale samples contained on the average 74% of the ^{131}I found in the fresh samples. As expected, kale with its large surface was the most sensitive of the samples to contamination. From precipitation data, the transfer factor for ^{131}I in kale was estimated at 5-10 pCi ^{131}I y · kg^{-1} per mCi ^{131}I km^{-2}, i.e., an order of magnitude more sensitive than milk but an order of magnitude less sensitive than grass for

which the transfer factor in 1976 was estimated at 50 pCi ^{131}I y kg^{-1} per mCi ^{131}I km^{-2}. As the annual mean consumption per caput of kale was 0.7 kg only, while the consumption of milk was 164 kg y^{-1} (table D.4.2.1.), the contribution of ^{131}I from vegetables was of the order of one tenth of that of milk (cf.3.2.3.).

2.5.4. Imported vegetable products

Denmark is almost self-sufficient as regards foodstuffs, but a few products are imported. Among these, tea and coffee contributed to the ^{90}Sr and ^{137}Cs content of the diet with (1.9 ± 1.2)% (1 SD) and (4.3 ± 2.5)%, respectively. Citrus fruit (oranges, lemons, grapefruit) and bananas constituted 1/4-1/3 by weight of the total fruit consumption in Denmark (RRD62), but imported fruit was in general of minor importance as a source of ^{90}Sr and ^{137}Cs in the diet; the mean contribution from citrus fruit and bananas to the total intakes of these radionuclides was (1.3 ± 1.1)% and (0.6 ± 0.4)%, respectively.

For the period 1963-1976 the mean contents of ^{90}Sr and ^{137}Cs in coffee were 10 and 44 pCi kg^{-1}, and for tea 45 and 259 pCi kg^{-1}, respectively. Tea thus contained levels approx. five times higher than those found in coffee. The concentrations were those actually extracted into the liquid coffee and tea, the activity remaining in coffee grounds and used tea leaves was thus excluded. One litre of water required 20 g tea or 50 g coffee (Da68), thus the liquid tea contained approx. twice as high concentrations of ^{90}Sr and ^{137}Cs than the liquid coffee. Neither the ^{90}Sr nor the ^{137}Cs showed significant variations with time, but the concentrations were in general higher in the first half of the sixties; this was especially evident for tea.

Citrus fruit sampled during 1963-1976 showed decreasing ^{137}Cs concentrations from approx. 25 to 3 pCi kg^{-1} and nearly constant ^{90}Sr levels at about 8 pCi kg^{-1}; bananas contained similar ^{137}Cs concentrations but the ^{90}Sr mean level was only 1.5 pCi kg^{-1}.

In dried fruit (raisins, dates, prunes and figs) the levels

decreased from approx. 100 pCi ^{137}Cs kg^{-1} in 1964 to approx. 10 pCi kg^{-1} in 1972, and from approx. 20 to 10 pCi ^{90}Sr kg^{-1}. The levels in nuts (almonds, walnuts, hazelnuts) decreased from approx. 500 pCi ^{137}Cs kg^{-1} to 50 pCi kg^{-1}, and the ^{90}Sr levels were around 50 pCi kg^{-1} during 1964-1972. Brazil nuts contained 7 times more ^{90}Sr than other nuts, compatible with a high stable Sr content in the Brazil nut of approx. 55 mg Sr kg^{-1}, or 30 mg Sr (g Ca)$^{-1}$. The ability to concentrate Sr as well as Ra (Ma58a) is connected to the content of polysaccharides in Brazil nuts. Brown algae also contain polysaccharides and they show a similar affinity to Sr (cf.2.7.1.).

It was not possible to propose meaningful prediction models for imported foods because the origin of the samples was unknown.

2.5.5. Faroese potatoes

A total area of approx. 1 km^2 grows potatoes in the Faroes and the annual production was approx. 1500 ts (Ri72) during the period of observation (table A.1.2.5.1.). The cultivation of other vegetables and fruit is of minor importance (Da58, Ka50), and most fruit and vegetables consumed in the Faroes are thus imported.

The variability among years of the ^{90}Sr as well as of the ^{137}Cs concentrations in Faroese potatoes was 0.44. In comparison, Danish potatoes showed a variability of 0.15 for ^{90}Sr and of 1.13 for ^{137}Cs. The Faroese data thus indicated a strong dependence on accumulated fallout in the case of ^{137}Cs, contrary to the Danish observations. Cæsium-137 in Faroese milk from Tvørå showed a low variability among years similar to that of the potatoes (cf.3.3.1.). The ability of potatoes to accumulate ^{137}Cs from Faroese soil makes potatoes one of the important ^{137}Cs donors in the Faroese diet, as they contribute 21 ± 12% (1 SD) of the total Faroese ^{137}Cs diet intake. The relative contribution from potatoes has been highest in recent years when the fallout rate has been low. As compared to the concentrations found in Danish potatoes during 1962-1976, Faroese potatoes contained 3.8 ± 1.9 (1 SD) times more ^{90}Sr and 43 ± 30 times more ^{137}Cs; the lowest ^{137}Cs ratios between the two

countries occurred in 1963-1964 when there were high fallout rates.

The prediction models for Faroese potatoes (tables C.2.5.1. - C.2.5.4.) indicated a radioecological sensitivity of 2.8 pCi $^{90}Sr\ kg^{-1}$ y per mCi $^{90}Sr\ km^{-2}$, or a little greater than that of Danish potatoes. For ^{137}Cs the sensitivity was 44 pCi $^{137}Cs\ kg^{-1}$ y per mCi $^{137}Cs\ km^{-2}$, or approx. 12 times higher than that of Danish potatoes; the radioecological sensitivities of grass (2.4.1.) and milk (3.2.2 and 3.3.2) from the two countries showed similar ratios, the sensitivity of grass being approx. 15 times and that of milk approx. 10 times higher in the Faroes.

The infinite time-integrated levels in Faroese potatoes originating from the ^{90}Sr and ^{137}Cs hitherto deposited in the Faroes became 426 pCi $^{90}Sr\ kg^{-1}$ y and 10.7 nCi $^{137}Cs\ kg^{-1}$ y.

2.6. Lichen

Og saa fløi Rensdyret afsted over Buske og Stubbe, gjennem den Store Skov, over Moser og Stepper alt hvad det kunde.
SNEDRONNINGEN

And the reindeer flew away across stubble and scrub, through the length of the forest, over bogs and prairies, as fast as he could go.
THE SNOW QUEEN

Lichens are long-lived (50-100 years) duplicate organisms composed of fungi and algae. As growths with persistent aerial parts and relatively large surface-to-weight ratios, lichens are likely to be effective accumulators of radioactive debris. Radioactivity measurements of lichens began in the late fifties (Go58, RRD58-59). It was, however, first when LIDÉN in 1961 (Li61) drew attention to the ^{137}Cs contamination of the arctic food chain - lichen-reindeer-man - that lichen became an object of primary radioecological interest.

In Greenland, reindeer live on the west coast, and they are most abundant in the district around Sukkertoppen (Vi71). Since the beginning of the sixties samples of lichens have been obtained from the west coast of Greenland and measured for ^{90}Sr and ^{137}Cs. Lichens have also been collected from other parts of Greenland, but these samples have not been considered in the present context.

2.6.1. Variation with time and location

The samples chiefly consisted of Cladonia species; Cetraria and Alectoria species were, however, also found in most samples. The samples received suggested that they had been collected from the upper part of the lichen carpet and thus corresponded to the lichen layer normally grazed by reindeer; but the exact sampling depth was not reported. The ^{90}Sr and ^{137}Cs concentrations in the lichen samples did not show significant local variations; fig.2.6.1.1. and fig. 2.6.1.2. suggest decreasing levels with time but the differences among years were not significant. The ^{137}Cs levels in 1962 were, however, probably lower than those in the remaining period.

The variability of ^{90}Sr among years (0.41) was probably higher than that of ^{137}Cs (0.27) (table B.2.6.1.) which is compatible with the field loss of ^{90}Sr in lichen being higher than that of ^{137}Cs (Ne66). The low variability of ^{137}Cs was typical for samples related to the accumulated deposit; however, the contamination of lichen was not indirect but direct and the low variability was thus due to the long residence time of ^{137}Cs in the lichen. The local variability of lichen from west Greenland was lower than that of grass (2.4.1.), due to the

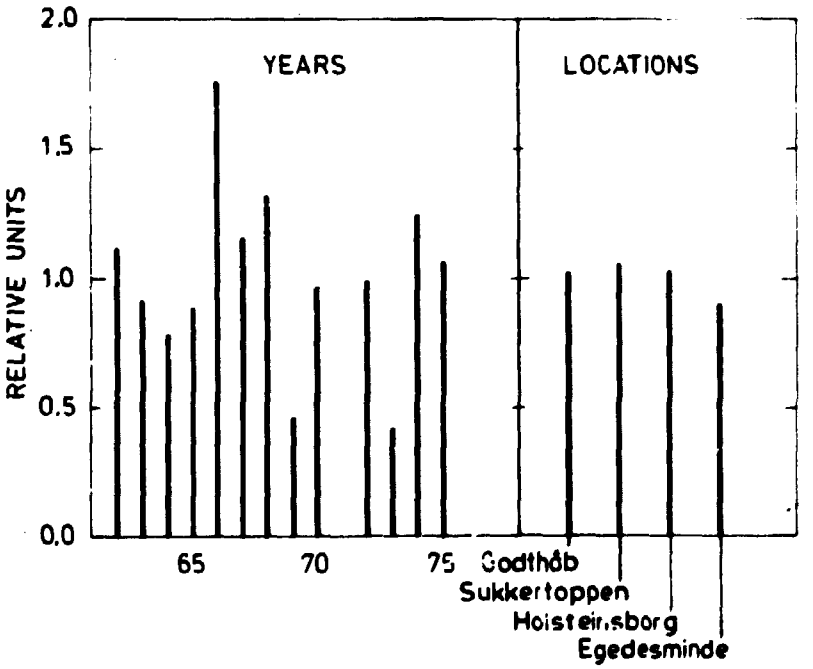

Fig. 2.6.1.1. The variation of nCi ^{90}Sr kg^{-1} in lichen collected in 1962-1975 in Greenland (cf. fig. A.1.1.3.3.). The bars show the concentrations relative to the grand mean 5.4 nCi kg^{-1} (= 1 at the relative scale).

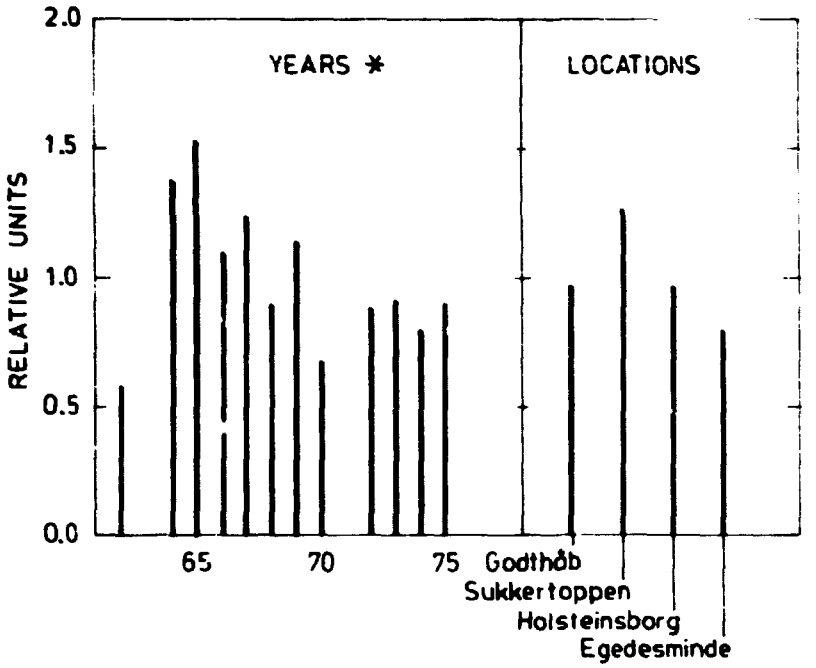

Fig. 2.6.1.2. The variation of nCi ^{137}Cs kg^{-1} in lichen collected in 1962-1975 in Greenland. The bars show the concentrations relative to the grand mean 25 nCi kg^{-1} (= 1 at the relative scale).

negligible radionuclide uptake from the soil in the case of lichen.

The most comprehensive studies of radionuclides in lichens have been carried out by PERSSON (Per70) and MATTSSON (Ma72) in Northern Sweden and by HANSON in Alaska (Han73). In the Swedish studies the annual mean ^{90}Sr levels varied between 5 and 13 nCi kg^{-1} and the ^{137}Cs levels between 16 and 50 nCi kg^{-1} during 1962-1970. For the same period, the Alaskan averages were 7.4 nCi ^{90}Sr kg^{-1} and 26 nCi ^{137}Cs kg^{-1}. The ^{90}Sr and ^{137}Cs concentrations in lichens were thus similar in Alaska, Lapland and Greenland. This was also expected from the similar fallout rates for the three territories, all of which are situated in the 60-70°N latitude belt. A more detailed inspection of the data showed that the $^{137}Cs/^{90}Sr$ ratios of the Alaskan lichen samples were in general lower than those of the Swedish and Greenlandic samples. This was because the Alaskan samples included the whole biomass, while the Greenlandic and Swedish samples only included the top layer of the plants. The Swedish studies have revealed decreasing $^{137}Cs/^{90}Sr$ ratios down through the lichen layer. Consequently, the Alaskan samples should contain relatively more ^{90}Sr than the other samples.

The effective half-lives of ^{90}Sr and ^{137}Cs in lichen have also been studied. MATTSSON thus estimated an effective half-life of 8-14 years for ^{137}Cs in undisturbed carpets of Cladonia alpestris, but in lichen carpets grazed by reindeer the half-life was only 5-6 years. HANSON concluded that an effective half-life of more than 10 years was reasonable for ^{137}Cs in lichens. In fig.2.6.1.2. the half-life of ^{137}Cs in West Greenland lichen during 1964-1975 is estimated at 14 years. PERSSON estimated the effective half-life of ^{90}Sr in the lichen carpet at 2.5 $\pm$ 0.8 years, and for lichen podetia HANSON experimentally found effective half-lives of 1.0 - 1.6 years and in field studies approx. 3 years. The ^{90}Sr in West Greenland lichen did not seem to decay in accordance with such short half-lives. The slope of the regression line corresponded to an effective half-life of 11 years. It should, however, be noted that the ^{90}Sr levels in lichen samples are rather sensitive to the sampling depth; as the Greenland samples were less well defined in this respect than the Swedish and Alaskan samples, it may be too early to draw any definite conclusions, although the period of observation in the present material was extended by five years as compared to the other two studies.

2.6.2. Prediction models

It was not possible to obtain empirical models for the ^{90}Sr concentrations in Greenland lichen for which the predicted values were significantly correlated with the observed values. The best fit showed a transfer factor of 1.6 nCi ^{90}Sr kg^{-1} y lichen per mCi ^{90}Sr km^{-2} deposited (table C.2.6.1.), and the estimated total infinite exposure integral in South-West Greenland from an estimated mean deposition of 55 mCi ^{90}Sr km^{-2} (the estimated mean deposition at Godthåb, cf.D.3.4.4.) was 88 nCi ^{90}Sr kg^{-1} y.

In the case of ^{137}Cs in lichen, the radioecological sensitivity was 17.6 nCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2}. The total infinite exposure integral from 88 mCi ^{137}Cs km^{-2} became 1550 nCi ^{137}Cs kg^{-1} y. The above transfer factors suggest that lichen was approx. an order of magnitude more sensitive to contamination with ^{137}Cs than with ^{90}Sr. NEVSTRUEVA et al. (Ne66) have experimentally shown that lichen concentrates ten times more

^{137}Cs than ^{90}Sr, when it was immersed in a water solution of the two nuclides. As compared with Greenland grass, lichen was approx. 50 times more sensitive to contamination with ^{137}Cs and approx. 3 times more sensitive to ^{90}Sr. NEVSTRUEVA et al. found that the upper parts of lichen contained 50-100 times more ^{137}Cs than contained in annual grasses.

The radioecological sensitivity of lichen is thus exceptionally high, and it is evident that links subsequent to lichen in the food chain are also expected to display high radioecological sensitivities.

2.7. Sea plants

Paa den opskyllede Tang laae elleve hvide Svanefjer;
DE VILDE SVANER

On the washed-up seaweed lay eleven white swan-feathers.
THE WILD SWANS

Phytoplankton concentrates several radionuclides from water and may thus be used as an indicator organism for radioactive contamination of the aquatic environment. Normally, sea plants are not part of the human food chain, but the well known porphyra-laverbread-man pathway of the Irish sea that transferred ^{106}Ru from the Windscale reprocessing plant to Welsh population groups is an exception (He76, Fo71). Among fission products, ^{90}Sr is especially concentrated by brown algae, and ^{90}Sr concentrations in sea water may thus be monitored by measuring the ^{90}Sr levels in sea plants.

Since 1957 samples of seaweed (Zostera marina), which is a higher plant, and bladderwrack (Fucus vesiculosus), which is a brown algae, have been collected every half-year from Roskilde Fjord.

2.7.1. Variation with time, species and location

Anovas showed a highly significant variation in the ^{90}Sr concentrations with time (fig.2.7.1.). The variability among years (table B.2.7.1.) was 0.64 for both species, i.e. higher than in sea water, which feature was ascribed to direct contamination through rain water when the sea plants were found

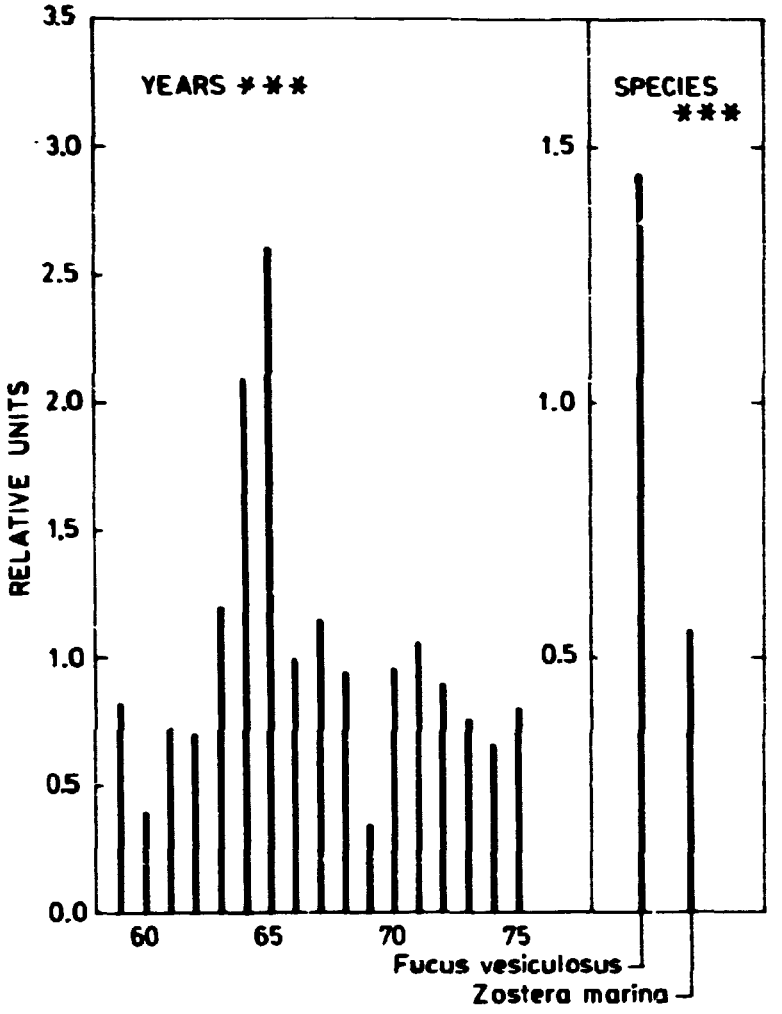

Fig. 2.7.1. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in sea plants collected in 1959-1975 in Roskilde Fjord (cf. figs. A.1.1.5. and A.1.2.4.). The bars show the levels relative to the grand mean 9.9 pCi (g Ca)$^{-1}$ (= 1 at the relative scales).

on the surface; the shallow water in Roskilde Fjord may have enhanced the direct contamination. The maximum occurred in 1965 and since then the levels have generally decreased; at the beginning of the seventies the ^{90}Sr concentrations were thus approx. 1/3 of the maximum. There was no significant difference between the ^{90}Sr concentrations in sea plants from the various seasons of the year, although the summer levels tended to be higher than the winter concentrations, which was the general trend also shown by Danish sea water (fig.1.5.1.).

The pCi ^{90}Sr (g Ca)$^{-1}$ levels in Fucus vesiculosus were 2-3 times higher than in Zostera marina, and there was no significant interaction between species and sampling years. In a study of the interspecific variation in ^{90}Sr concentration in

sea plants (RRD61), it appeared that the ^{90}Sr (g Ca)$^{-1}$ ratios were correlated with the stable Sr to Ca ratio of the plants. The stable Sr to Ca ratio in Fucus vesiculosus was thus 3 times higher than in Zostera marina (RRD59-76), and Fucus serratus and Laminaria digitata contained approx. twice as high mg Sr (g Ca)$^{-1}$ levels than Fucus vesiculosus, while Furcellaria fastigiata (a red algae) only contained approx. 1/4 of the levels found in Fucus vesiculosus (RRD61). Brown algae and especially Fucus serratus and Laminaria digitata thus concentrate Sr from sea water. As compared to the mg Sr (g Ca)$^{-1}$ ratios in sea water, these two species contained 4 times higher ratios than the water (Pet62), i.e. they preferred Sr 4 times as much as Ca. It has been shown that alginates rich in guluronic acid have a greater affinity with Sr in the ion-exchange reaction strontium-calcium (Ha67). Because brown algae in particular, such as Laminaria, are relatively rich in guluronic acid this explains why these species concentrate Sr in preference to Ca from sea water.

Brown algae - mainly Laminaria and Fucus species - have also been obtained from Faroese and Greenlandic waters. The ^{90}Sr concentrations in these samples were generally lower than in the corresponding Danish samples, as expected from the lower ^{90}Sr levels in Faroese and Greenlandic sea water (1.5.2.). However, the sea plants contained relatively more ^{90}Sr than the low ^{90}Sr concentrations in Faroese and Greenlandic waters might have suggested. This was partly because the samples taken consisted of Laminaria and Fucus serratus, which as shown above concentrate Sr more effectively than Fucus vesiculosus. However, the most important reason for the enhanced levels was undoubtedly direct contamination through precipitation of the plants occurring on the surface. This method of exposure was relatively more important in the low activity Faroese and Greenlandic waters.

2.7.2. Relations and prediction models

Fucus vesiculosus contains approx. 10 g Ca per kg wet weight, the dry matter content being 20%. As the stable Sr content is approx. 30 mg Sr (g Ca)$^{-1}$, (RRD59-76) 1 kg bladderwrack (wet

weight) contains 300 mg Sr. The concentration of Sr measured in the Sound is 2.4 mg Sr l^{-1} (salinity 13.4‰)(Pet62), hence the expected concentration factor (CF) between bladderwrack and sea water becomes 125, i.e. 1 kg bladderwrack at equilibrium is supposed to contain the amount of Sr present in 125 l of sea water.

The prediction models for sea plants (table C.2.7.1.) show that the radioecological sensitivity was 2.7 for Zostera marina and 10.7 pCi ^{90}Sr (g Ca)$^{-1}$ y per mCi ^{90}Sr km^{-2} for Fucus vesiculosus; the higher sensitivity for Fucus was in agreement with the higher mg Sr (g Ca)$^{-1}$ quotient in this species. When the Fucus value was combined with the radioecological sensitivity for Danish sea water (1.5.1.), the transfer factor from sea water to Fucus vesiculosus was estimated at $\frac{10.7 \cdot 10}{0.35}$ = 306 pCi ^{90}Sr kg^{-1} y per pCi ^{90}Sr l^{-1} y. This value was higher than that estimated for stable Sr above because, as mentioned above, the direct contamination with ^{90}Sr came into play for the airborne fallout ^{90}Sr. In the case of a pure waterborne release of ^{90}Sr a closer agreement would have been expected between the radioactive and the stable Sr CF.

The sampling of sea plants in the Faroes and in Greenland was less systematic than in Denmark. The prediction models (table C.2.7.1.) showed radioecological sensitivities lower than for Fucus in Denmark. In the Faroes the transfer factor was estimated at 1.2 pCi ^{90}Sr (g Ca)$^{-1}$ y per mCi ^{90}Sr km^{-2} and in West Greenland at 5.6. As compared with sea water (1.5.2.), the transfer was 38 pCi ^{90}Sr (g Ca)$^{-1}$. y per pCi ^{90}Sr l^{-1} y in the Faroes and 24 in West Greenland, as compared with 30 in Denmark for Fucus. Considering instead the "observed ratios" between the pCi ^{90}Sr (g Ca)$^{-1}$ quotients in sea plants and in sea water, these become 16 for the Faroes, 8 for Greenland and 6 for Denmark, which demonstrates the relatively greater importance of direct contamination in the low activity waters.

2.8. Conclusions

2.8.1. General

Apart from the influence of radionuclide concentrations in the abiotic environment on the radioactive contamination of vegetation, environmental factors such as climate, soil type and agricultural practice also play an important modifying role. These factors affect the radioecological sensitivity as well as the variability of the radionuclide levels in vegetation.

Crops with a large surface-to-weight ratio and slow growth show in general higher direct contamination than plants of an opposite nature. Indirect contamination is an important pathway for ^{90}Sr in many vegetables. In the Faroes indirect contamination is mainly responsible for the enhanced ^{137}Cs levels in vegetation. Lichen holds an exceptional position due to its high radioecological sensitivity, especially to ^{137}Cs contamination.

2.8.2. Cereal grain

The variability among years was greater for ^{137}Cs than for ^{90}Sr in grain. In the case of ^{137}Cs, the variability ($CV_{p\ years}$ = 1.8) was similar to that in precipitation and air, in agreement with expectations, because the ^{137}Cs was derived from direct contamination only. Rye and barley showed higher variabilities among years for ^{90}Sr ($CV_{p\ years}$ = 1) than did wheat and oats ($CV_{p\ years}$ = 0.7) because of the higher indirect contamination of the latter.

The variability among species was lower for ^{90}Sr than for ^{137}Cs because indirect contamination counteracted direct contamination in the case of ^{90}Sr. Plutonium showed a marked interspecific variability, barley showing 5-6 times higher concentrations than wheat.

The local variability was more pronounced for ^{90}Sr ($CV_{p\ locations}$ = 0.4) than for ^{137}Cs ($CV_{p\ locations}$ = 0.2); for ^{90}Sr, it was

lower for rye than for the other species. This also reflected the differences between the two nuclides and among species with respect to indirect and direct contamination.

The radioecological sensitivities were 32 pCi $^{90}Sr\ kg^{-1}$ y per mCi $^{90}Sr\ km^{-2}$ in the case of rye, 26 for barley, 24 for wheat and 37 for oats; in the case of ^{137}Cs the sensitivity was 46 pCi $^{137}Cs\ kg^{-1}$ y per mCi $^{137}Cs\ km^{-2}$ for rye, 32 for barley, 28 for wheat and 27 for oats. Jutland showed 1.4 and 1.2 times higher radioecological sensitivities than the Islands regarding ^{90}Sr and ^{137}Cs contamination, respectively.

Manganese-54 occurred in grain in 1963-1965; the radioecological sensitivity of grain to ^{54}Mn contamination was approx. 75% of that of ^{137}Cs, and the contamination was direct.

2.8.3. Bread

The local variability of ^{90}Sr and ^{137}Cs in Danish bread was lower than that of grain because rye was transferred from Jutland to the Islands, while the opposite was the case for wheat. The mean ratios between the concentrations of the two radionuclides in bread and grain were 0.75 for rye-bread - rye for both nuclides, and in the case of white-bread - wheat 0.15 for ^{90}Sr and 0.4 for ^{137}Cs. Manganese-54 showed a ratio for rye bread similar to that of ^{90}Sr and ^{137}Cs, but the ^{54}Mn concentration in white bread was only 9% of the concentration in wheat. Danish rye bread was thus an exceptionally important source of ^{54}Mn among cereal products.

Faroese bread contained half as much ^{90}Sr as Danish bread and 60-90% as much ^{137}Cs, because the degree of extraction of Faroese rye flour is less than that of Danish rye flour, and also because more than half of the Faroese flour is imported from countries other than Denmark. The creta praeparata in Danish cereal products contributed nearly 40% of the total calcium intake with the adult human diet. The amount of calcium in bread varied within the country, bread from Lolland-Falster thus contained more calcium than bread from West Jutland. Bread from the Faroes contained half as much creta praeparata as Danish bread.

2.8.4. Grass and other fodder crops

The local variability of ^{90}Sr in Danish grass ($CV_{p\ locations}$ = 0.2) was similar to that of the deposition of ^{90}Sr, but ^{137}Cs showed a higher local variability. In general, the first quarter of the year displayed the highest ^{90}Sr levels in grass.

The radioecological sensitivity of Danish grass was 31 pCi ^{90}Sr $(g\ Ca)^{-1}$ y per mCi ^{90}Sr km^{-2}; the value for Faroese grass was 216, and for Greenlandic grass 442. In the case of ^{137}Cs, Danish grass had a sensitivity of 5 pCi ^{137}Cs $(g\ K)^{-1}$ y per mCi ^{137}Cs km^{-2}, Faroese 76 and Greenlandic 79. The high sensitivities in the Faroes and Greenland were ascribed to the pronounced indirect contamination of grass in these areas.

The $^{137}Cs/^{90}Sr$ ratio of grass and hay was higher than that of fodder from roots and silage of root leaves. This phenomenon influenced local as well as seasonal variations of ^{90}Sr and ^{137}Cs in cows' milk in Denmark.

2.8.5. Vegetables and fruit

The variability among years of ^{90}Sr in cabbage, carrots and potatoes ($CV_{p\ years}$ = 0.2) was similar to that observed for the accumulated fallout in soil, indicating a strong dependence on indirect contamination. The variability of ^{137}Cs, on the other hand, ($CV_{p\ years}$ = 1.2) suggested direct contamination as the main pathway. Apples displayed a relatively high variability among years ($CV_{p\ years}$ = 1.1) for ^{90}Sr as well as for ^{137}Cs, also an indication of direct contamination.

The local variabilities ($CV_{p\ locations}$ = 0.2) were similar for ^{90}Sr and ^{137}Cs and close to the variability of ^{90}Sr deposition. The local variability of ^{90}Sr in vegetables was lower than that observed in grain. Higher stable Sr/Ca ratios in vegetables from the eastern part of the country suggested enhanced indirect contamination at the low fallout stations, which would reduce the local variability.

Leaf vegetables showed higher pCi ^{90}Sr kg^{-1} than root vegetables because of a higher calcium content and greater direct con-

tamination. On the other hand, the stable Sr/Ca ratios in leaf vegetables were two-thirds of those in roots, which suggested enhanced indirect contamination of roots relative to leaf vegetables.

The radioecological sensitivity of vegetables to contamination with ^{90}Sr was in general higher than that of ^{137}Cs, except for potatoes and apples, where the translocation of ^{137}Cs enhanced these levels relative to the less mobile ^{90}Sr. The radioecological sensitivity of vegetables was nearly the same in Jutland and in the Islands.

The radioecological sensitivity of Faroese potatoes was 44 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2}, while Danish potatoes showed a sensitivity of 3.8. In the case of ^{90}Sr the radioecological sensitivity was nearly the same for Danish and for Faroese potatoes ($\sim$ 2.7 pCi ^{90}Sr kg^{-1} y per mCi ^{90}Sr km^{-2}).

Coffee, tea, and imported fruits contributed only a few per cent to the total intakes of ^{90}Sr and ^{137}Cs.

2.8.6. Lichen

The variability of ^{90}Sr and ^{137}Cs among years ($CV_{p\ years}$ = 0.4 and 0.3, respectively) in Greenland lichen was relatively low suggesting that the lichen retained the radionuclides and that the field loss thus was low. The effective half-lives in the Greenland lichen were estimated at 14 years for ^{137}Cs and 11 years for ^{90}Sr.

The radioecological sensitivity of lichen was 18 nCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2}, which was higher than observed for any other sample in this study. The sensitivity to ^{90}Sr was approx. 10 times less than that of ^{137}Cs.

2.8.7. Sea plants

The time variability of ^{90}Sr in Fucus vesiculosus (a brown algae) and in Zostera marina (a higher plant) ($CV_{p\ years}$ = 0.6)

was higher than that observed in Danish inner waters, indicating some direct contamination of the plants with rain water. Fucus contained nearly 3 times more $^{90}Sr/Ca$ than Zostera, in agreement with the stable Sr/Ca ratios between the two species.

The "observed ratio" between brown algae and sea water was 4 for stable Sr/Ca, and the concentration factor between Fucus and sea water was 125 (g Sr kg^{-1} fresh weight per g Sr l^{-1} sea water), but for ^{90}Sr the factor was 3o6 because of the direct ^{90}Sr contamination of the sea plants.

The radioecological sensitivity of Fucus from Roskilde Fjord was 11 pCi ^{90}Sr $(g\ Ca)^{-1}$ y per mCi ^{90}Sr km^{-2} and for Zostera it was 3. In the Faroes and Greenland the radioecological sensitivities of sea plants were lower than in Denmark, but the transfer factors of ^{90}Sr from sea water to sea plants were higher as a result of the relatively higher direct contamination of marine vegetation through rain water in these areas.

3. MILK AND OTHER ANIMAL PRODUCTS (the consumers)

3.1. Introduction

The trophic level of the human food chain following the producers is made up of the consumers, i.e. animals and their products. The consumers utilized by man are mostly herbivores. This applies generally to domestic animals, although they may receive minor amounts of additional fodder of animal origin e.g. bone meal and fish meal. As regards the aquatic animals included in the human food chain, these are often carnivores and as such they may display enhanced levels of certain radionuclides, e.g. ^{137}Cs, compared to aquatic herbivores. Among the various animal products, milk is an important constituent of the human diet, at least in the industrialized world, as it is the main diet for infants (0-1 year) and because some of the most abundant fallout nuclides are easily secreted in milk. Therefore the emphasis in this chapter is laid on milk. Because Faroese milk holds an exceptional position with respect to radioactive contamination, a special section is devoted to the milk from this area. The section on terrestrial animals comprises Greenland reindeer in addition to domestic animals. The reindeer are expected to show a high radioecological sensitivity according to the high contamination levels found in lichen. Marine fish from Danish, Faroese and Greenlandic waters are dealt with in the following section, and the chapter is concluded with marine mammals, seabirds and hens' eggs.

3.2. Danish milk

"Gud ske lov for Melken!" sagde Konen, "nu kan vi faae Melkemad, Suur og Ost paa Bordet. Det var et deiligt Byttе!"
HVAD FATTER GJØR

"Thank goodness for some milk!" said the wife; "now we can have milk-puddings and butter and cheese to eat. What a lovely exchange!"
DAD'S ALWAYS RIGHT

Milk is an important constituent of European, North American and Oceanian diet. In modern society where breast-feeding of infants

is to a large extent supplemented or even replaced by cows' milk, this substance is the main source of diet for infants in their first year of life.

Fission products such as ^{89}Sr, ^{90}Sr, ^{131}I and ^{137}Cs are all readily transferred to milk. As these radionuclides are to a considerable extent absorbed in the gastrointestinal tract (Ic59), they contribute to the internal radiation dose of milk consumers if the fodder of the cows is contaminated by fission products.

Hence milk was included at an early stage in the environmental studies of radioactive contamination from nuclear weapons tests. A sample of cheese collected in Wisconsin in July 1953 was among the very first items to be measured for ^{90}Sr in the human environment. This sample contained 0.5 pCi ^{90}Sr (g Ca)$^{-1}$ (Ku62). In April 1954 the Health and Safety Laboratory (HASL) of the USAEC (Has58) initiated the first systematic study of ^{90}Sr in milk. The first samples consisted of powdered milk from Perry in New York State; a few months later this was supplemented by samples of liquid milk from New York City. In 1956 the Los Alamos Scientific Laboratory (Has58) began ^{137}Cs determinations in milk. The first attempt to measure ^{131}I fallout in milk was made in the USA in February-June 1955 parallel with a study of ^{131}I in the thyroid glands of cattle and of humans (Co57a). In the last half of the fifties studies of fallout nuclides in milk were rapidly expanded because of the increasing intensity of nuclear weapons tests, and also because of improved analytical techniques. Data from most areas of the world have since 1958 been compiled in the UNSCEAR reports (Un58-77). The first Danish data on ^{90}Sr and ^{137}Cs in milk appeared in 1959 (RRD59). The samples came from four dried milk factories selected for this purpose by Risø in co-operation with the National Health Service of Denmark.

Throughout the years, the ^{90}Sr and ^{137}Cs contents of Danish cheese have occasionally been measured (RRD59-76). However, it has been shown (Re66) that both the ^{90}Sr/Ca and the ^{137}Cs/K ratios in general seem unaffected by dairy processes. The ^{90}Sr and ^{137}Cs concentrations in the various dairy products

may therefore be calculated from the milk analysis, if the concentrations of Ca and K in the products are known. In the case of cheese, it has been assumed in the total diet estimates (4.2.) that 1 kg cheese contains 8.5 g Ca and 1.2 g K.

3.2.1. Variation with time and location

The ^{90}Sr and ^{137}Cs concentrations in milk have shown a significant variation among years (tables B.3.2.1 and B.3.2.2, figs.3.2.1.1, and 3.2.1.2). The maximum occurred in the last half of 1963, and since then the levels have generally been decreasing, ^{90}Sr more slowly than ^{137}Cs, in agreement with the observations for vegetation. The variability among years was 0.62 for ^{90}Sr and 1.02 for ^{137}Cs (1961-72); which are lower variabilities than found for grass (2.4) and grain (2.2), but higher than those of vegetables (2.5). The variation among years depended primarily on the fallout rate and, for ^{90}Sr, also on the accumulated fallout in the soil. However, these were not the only reasons for variation. The composition of cattle fodder has thus been altered during the period in question. In 1957 the ratio between feed units from roots and grass was 1.0, while it had decreased to approx. 0.65 by the beginning of the seventies (Da57-77). From table 2.4.2. it appears that this implies a relatively slower annual decrease in the ^{137}Cs levels than in the ^{90}Sr concentrations of the milk. Another factor that has interferred with the general fall in levels has been the more rapid decrease in milk production in the Islands than in Jutland (Da57-77). This has enhanced the relative mean concentration of ^{90}Sr and especially of ^{137}Cs in Danish milk, because milk from Jutland shows higher levels than milk from the Islands (figs.3.2.1.1. and 3.2.1.2.).

Beside the annual variation, the seasonal variation within the year was studied. This variation was most pronounced in the case of ^{137}Cs; the variability among months was thus 0.34 for ^{137}Cs, while it was only 0.04 for ^{90}Sr. If the monthly variations within the so-called "milk year" ($May_{(i)}$ - $April_{(i+1)}$) are considered, i.e. the period in which the fodder used for milk production originates mainly from the year (i), the ^{90}Sr levels were nearly constant from month to month. The ^{137}Cs

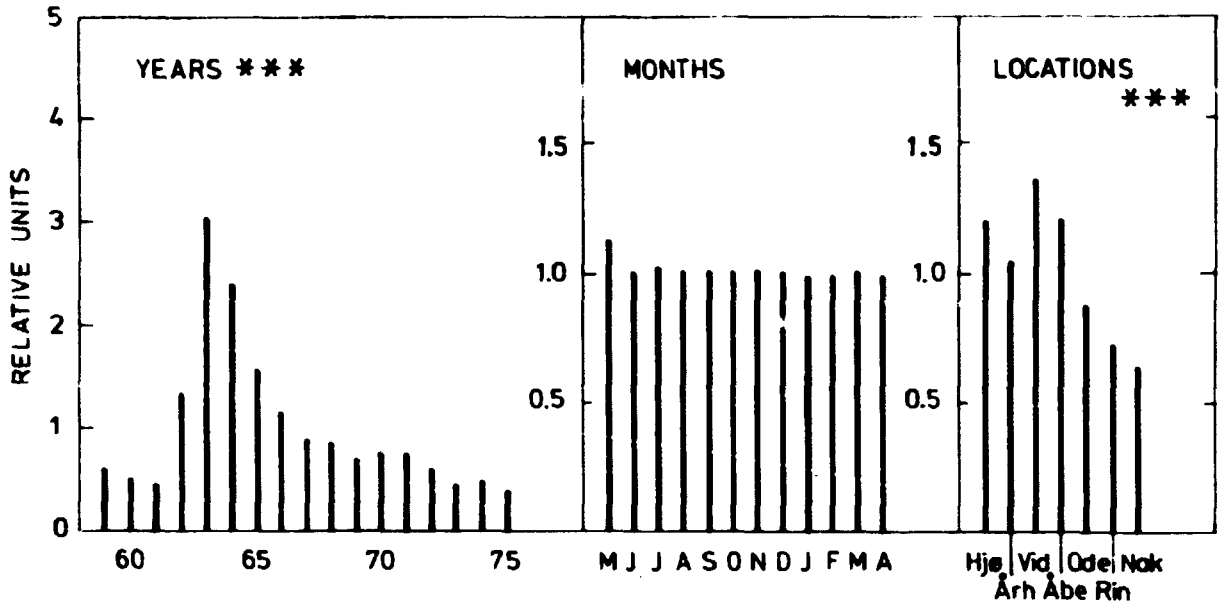

Fig. 3.2.1.1. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in Danish dried milk collected monthly at 7 dried milk factories (cf. Fig. A.1.3.2.) from May 1959 to April 1976 (cf. Table A.1.3.2.). The bars indicate the levels relative to the grand mean 9.4 pCi (g Ca)$^{-1}$ (= 1 at the relative scales). The years were "milk years" (cf. the text).

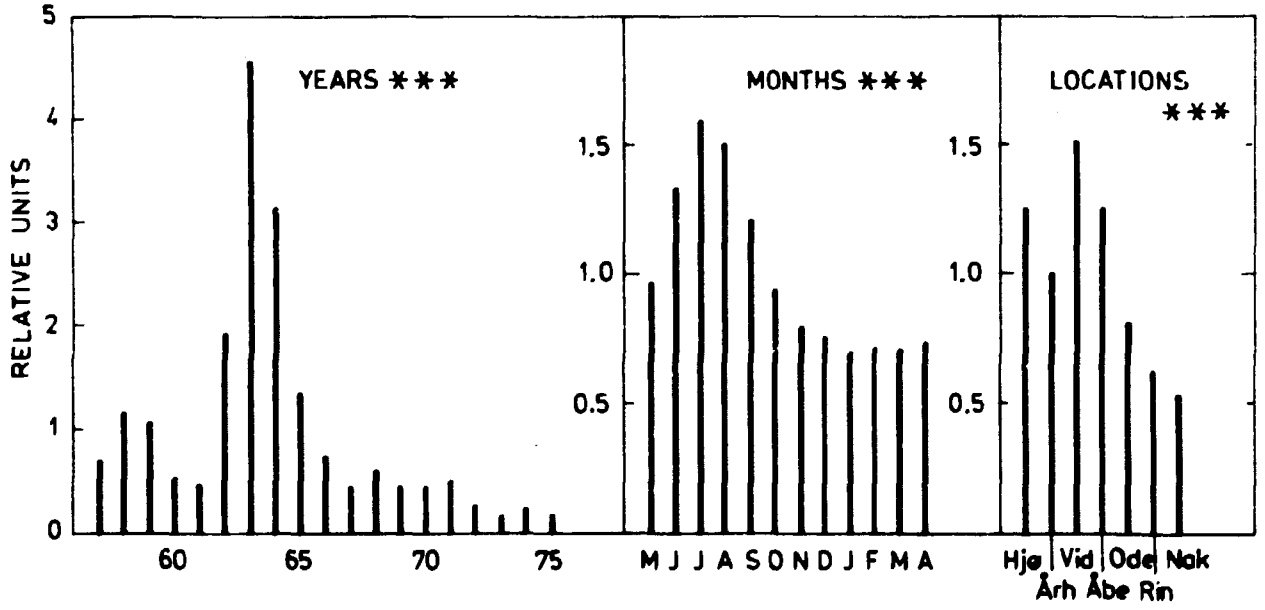

Fig. 3.2.1.2. The variation of pCi ^{137}Cs (g K)$^{-1}$ in Danish dried milk collected monthly at 7 dried milk factories (cf. Fig. A.1.3.2.) from May 1959 to April 1976 (cf. Table A.1.3.2.). The bars indicate the levels relative to the grand mean 18.5 pCi (g K)$^{-1}$ (= 1 at the relative scales). The years were "milk years" (cf. the text).

concentrations were, however, 2-3 times higher in July than in January. The $^{137}Cs/^{90}Sr$ ratio of the summer milk was thus significantly higher than that of the winter milk. This seasonal variation was a result of the seasonal change in the composition of the fodder of the cows. It appears from table

2.4.2. that the $^{137}Cs/^{90}Sr$ ratio in summer fodder was significantly higher than in winter fodder.

Anovas showed a significant interaction between "milk years" and months. During periods with increasing fallout levels (1961-1963), the autumn months showed relatively high concentrations of ^{90}Sr and ^{137}Cs in the milk because of the contamination of grass and root vegetable leaves by fresh fallout (from the autumn test series in 1961 and 1962). During periods with decreasing levels of contamination (1964-1967), the relatively high milk concentrations observed in the late spring were due to the consumption by the cows in May of some residual fodder (from the preceding "milk year").

The local variations of the ^{90}Sr and ^{137}Cs concentration in Danish milk were similar for the two nuclides (figs.3.2.1.1. and 3.2.1.2.). The highest levels were observed in the western parts of the country where also the highest fallout levels and thus the highest concentrations in the crops occurred. As was the case for the time variation, ^{137}Cs showed a more pronounced local variation than ^{90}Sr, which was in contrast to the observations made for, e.g., grain (2.2.). The local variability (table B.3.2.3.) was 0.39 for ^{137}Cs and 0.26 for ^{90}Sr. The quotient between the ^{137}Cs concentrations in milk from Videbæk in West Jutland and Nakskov in Lolland-Falster was 3.0, while for ^{90}Sr it was 2.2. In other words, the $^{137}Cs/^{90}Sr$ ratio of milk from Nakskov was lower than that of milk from Videbæk. This difference was due to differences in the composition of the fodder in the two areas. The ratio between feed units produced from roots and offal (from sugar beet production) and from grass was nearly twice as high in Lolland-Falster than in West Jutland (table D.3.2.1.). The cattle-feed in Lolland-Falster thus contained relatively more roots and less grass than the fodder used in the other parts of the country (Hau73). Table 2.4.2. indicates that a high root-to-grass ratio resulted in a lower $^{137}Cs/^{90}Sr$ ratio than in diets with a low root-to-grass ratio. STEWARD et. al (Ste65) have shown that the transfer of ^{137}Cs from cattle feed to milk was closely related to the crude fiber content of the feed:

the higher the crude fiber content, the lower the percentage of ^{137}Cs intake secreted per litre of milk. As the crude fiber content of grass is higher than that of roots (Hau73), one would expect a relatively lower secretion of ^{137}Cs in milk from cows on a grass diet than in milk from cows on a root diet. The lowering effect of a high root/grass ratio in cattle feed on the ^{137}Cs levels in milk may therefore be counteracted but not necessarily compensated by an increasing effect on the ^{137}Cs levels from the lower crude fiber content of such feed.

Anovas of the quotients between the ^{90}Sr concentrations in milk from Videbæk and in milk from Nakskov showed no significant variation within the "milk year", but the ratio increased through the years. This was ascribed to the increasing importance of root uptake of ^{90}Sr, which played the greatest role for the sandy soils in West Jutland (Videbæk) (An67a, An67b) (cf. also the greater soil factors in the prediction models for crops from Jutland (e.g. table C.2.2.1.)).

The interactions between time and location were in general not very pronounced; the local pattern of the ^{90}Sr as well as of the ^{137}Cs levels in the milk was nearly unchanged with time. Besides the above-mentioned influence of variations in the relative importance of direct to indirect contamination from one place to another, yearly alterations in the relative feed habits of the cattle for the various areas, and shifts in the livestock supplying the milk to a given factory, should be considered. It is remarkable that the interaction between locations and years was less pronounced for ^{137}Cs, which in Danish milk generally depends on direct contamination. This suggests that variations in root uptake were mainly responsible for the interaction observed for ^{90}Sr. A study of the monthly pattern of the radionuclide concentrations in the milk at the individual locations did not reveal any marked variations in the pattern for ^{90}Sr from one place to another. In the case of ^{137}Cs, Videbæk and Nakskov showed a marked and narrow peak in July, whereas Hjørring and Åbenrå showed a broader maximum covering July-September. Århus, Odense and Kalundborg held an intermediate position. A broad maximum suggested a relatively constant application of additional fodder throughout the grazing season,

whereas a narrow peak reflected decreasing amounts of additional fodder throughout the grazing season until July, and thereafter increasing amounts.

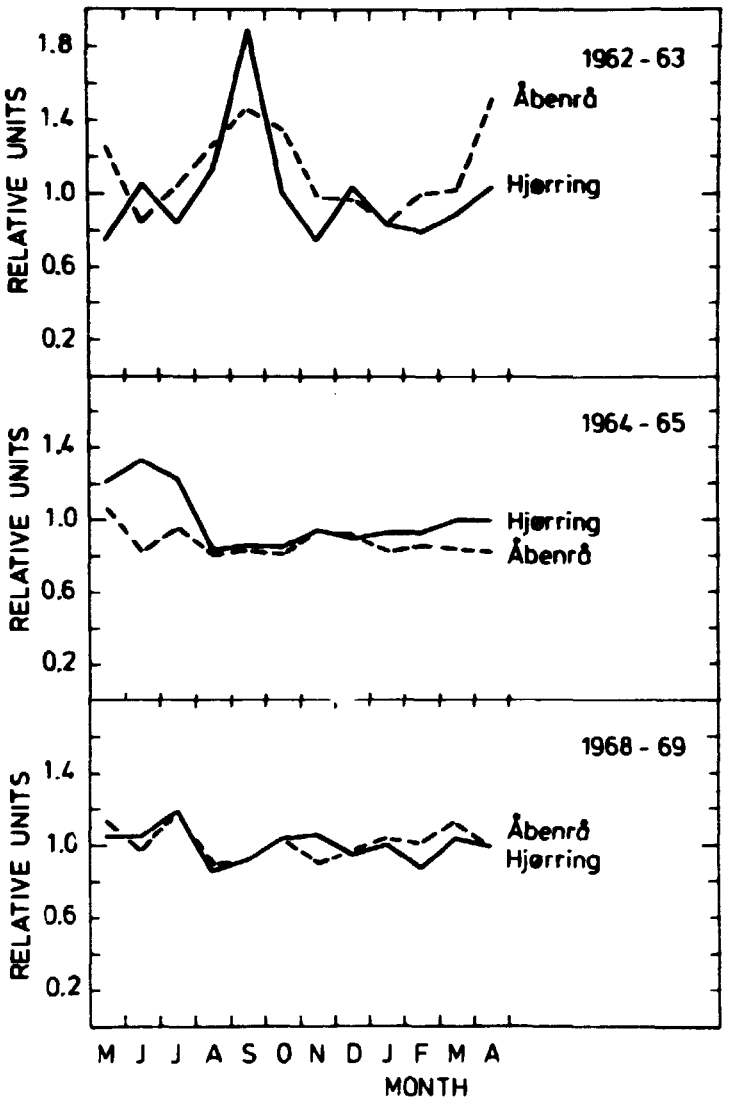

Fig. 3.2.1.3. An illustration of first- (L×M, Y×M, Y×L) and second-order (Y×M×L) interactions (Table B.3.2.1.) of pCi ^{90}Sr (g Ca)$^{-1}$ in Danish dried milk. Where there were no interactions, the curves for the two localities should have been parallel both within and among milk years.

The second-order interaction between years, locations and months was significant for ^{90}Sr in Danish dried milk. Figure 3.2.1.3. exemplifies this interaction. It appeared that the first-order interactions between locations and months varied among the years. As for grain (2.2.1.), a main reason for the interactions observed in milk was variations in the relative importance of direct and indirect contamination with time and location; but

furthermore the interactions for milk were influenced by the variations in the composition of the cattle feed.

An anova of the quotients between dried milk and fresh milk of pCi ^{90}Sr (g Ca)$^{-1}$ did not reveal any significant time variation between the two types of milk, but the local variation was significant. The dried milk from Videbæk thus showed higher concentrations than the fresh milk from West Jutland, while dried milk from Nakskov showed lower levels than fresh milk from Lolland-Falster. The local variability of ^{90}Sr in dried milk ($CV_{p\ locations} = 0.26$) was a little higher than that of fresh milk ($CV_{p\ locations} = 0.22$); in the case of ^{137}Cs, the local variabilities of the two milk types were 0.39 and 0.27, respectively (table B.3.2.3). The countrywide mean quotient between the pCi ^{90}Sr (g Ca)$^{-1}$ levels in dried milk and fresh milk was 1.09 $\pm$ 0.22 (1 SD) (231 ratios), and for pCi ^{137}Cs (g K)$^{-1}$ the mean ratio was 1.22 $\pm$ 0.48 (227 ratios); the ^{137}Cs ratio was generally higher in summer than in winter. The dried milk factories thus apparently received some of their milk from areas with a higher milk concentration of ^{90}Sr and ^{137}Cs than the consumption average for the areas in which the factories were situated. The higher $^{137}Cs/^{90}Sr$ ratios in the dried milk suggested that the suppliers of milk to the factories other than the local were situated in the western part of the country, because the $^{137}Cs/^{90}Sr$ ratio in milk from Jutland was somewhat higher than in milk from the Islands. The fresh milk samples made it furthermore possible to evaluate the production and the population-weighted countrywide activity means in Danish milk (RRD59-76). The mean ratio between the two weighted means was approx. 1.11. This showed that the milk used for consumption in Denmark as fresh milk had a lower activity content than the mean activity of the total production. The Danish population thus obtained a reduction in its intake of ^{90}Sr and ^{137}Cs from milk by preferentially using the more contaminated milk to other milk products such as butter and cheese, which are exported to a large extent. As most milk was produced in Jutland*), it agrees with expectations that the surplus pro-

*) At the beginning of the sixties two-thirds of all milk produced in Denmark came from Jutland, ten years later the fraction had increased to four-fifths (Da57-77).

duction mainly originated from this area, and thus showed a higher mean content of ^{90}Sr and ^{137}Cs than the country mean, which was not significantly different from the population-weighted mean (RRD59-76). However, since the beginning of the seventies, the latter mean has shown a tendency to increase because of the transfer of milk from Jutland to the eastern part of the country, especially to Copenhagen.

The summer sampling of untreated whole milk from livestock at the state experimental farms during 1962-1970 showed that the mean concentrations in this milk were somewhat lower than those of dried milk, just as observed above for the fresh consumers milk. The mean ratios between dried milk and whole milk were 1.06 ± 0.27 (1 SD) (73 ratios) for ^{90}Sr and 1.28 ± 0.62 for ^{137}Cs (68 ratios).

3.2.2. Relations and prediction models

The prediction models for milk have been throughly studied by many investigators. Already in 1958 in the first UNSCEAR report (Un58) a model was proposed for the ^{90}Sr concentration in milk. This model assumed the milk concentration of ^{90}Sr to depend on the fallout rate of ^{90}Sr and on the ^{90}Sr accumulated in the soil. Later, more sophisticated models were suggested and these included a so-called lag rate factor (Ba66, Aa66a) which takes the fallout rate in the preceding year into acount. These models were further refined by assuming effective half-lives of the ^{90}Sr in milk of less than the physical half-life of 28 years (Ba72, Be72). Figures 3.2.2.1.-3.2.2.4. show the most recent prediction models for ^{90}Sr and ^{137}Cs in milk for the Islands and Jutland compared with the observed values. Dried milk was more sensitive to contamination by ^{137}Cs than by ^{90}Sr and milk from Jutland was more sensitive to contamination than milk from the Islands (tables C.3.2.1 and C.3.2.2.).

The radioecological sensitivity of Danish milk was estimated from the two independent transfer factor estimates based on dried milk and fresh milk, respectively. If all equations in tables C.3.2.1. and C.3.2.2. are used, the means of the four estimates for the entire country of each nuclide are 3.25 $\pm$

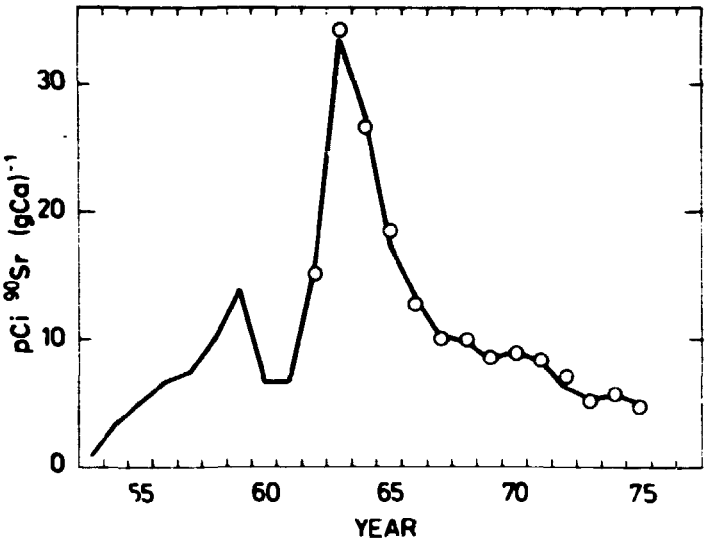

Fig. 3.2.2.1. Predicted (the curve calculated from Table C.3.2.1. No. 1) and observed pCi ^{90}Sr (g Ca)$^{-1}$ levels in dried milk from Jutland. Correlation coefficient r = 0.9979***.

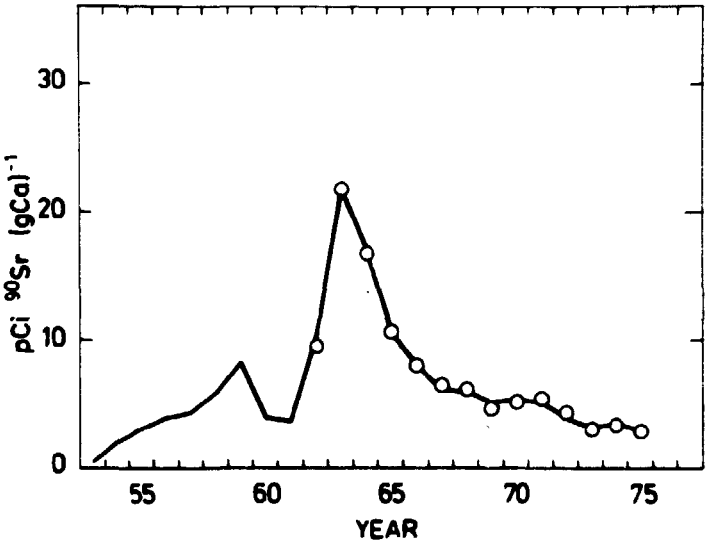

Fig. 3.2.2.2. Predicted (the curve calculated from Table C.3.2.1. No. 3) and observed pCi ^{90}Sr (g Ca)$^{-1}$ levels in dried milk from the Islands; r = 0.9977***.

0.23 (1 SE) pCi ^{90}Sr (g Ca)$^{-1}$ y per mCi ^{90}Sr km^{-2} and 3.43 ± 0.27 pCi ^{137}Cs (g K)$^{-1}$ y per mCi ^{137}Cs km^{-2}. The present transfer factor for ^{90}Sr is 4% higher than the transfer factor most recently estimated for Danish milk by UNSCEAR (Un77); but in the case of ^{137}Cs the UNSCEAR estimate was 27% higher than the above value. The UNSCEAR models assumed lower root uptake of ^{90}Sr but higher of ^{137}Cs than the present models.

The present estimate of the radioecological sensitivity of ^{90}Sr was only half of the value proposed in 1968 (X) when the effective half-life of ^{90}Sr available for indirect contamination was assumed equal to 28 years; the estimate for ^{137}Cs was 71% of the 1968 value.

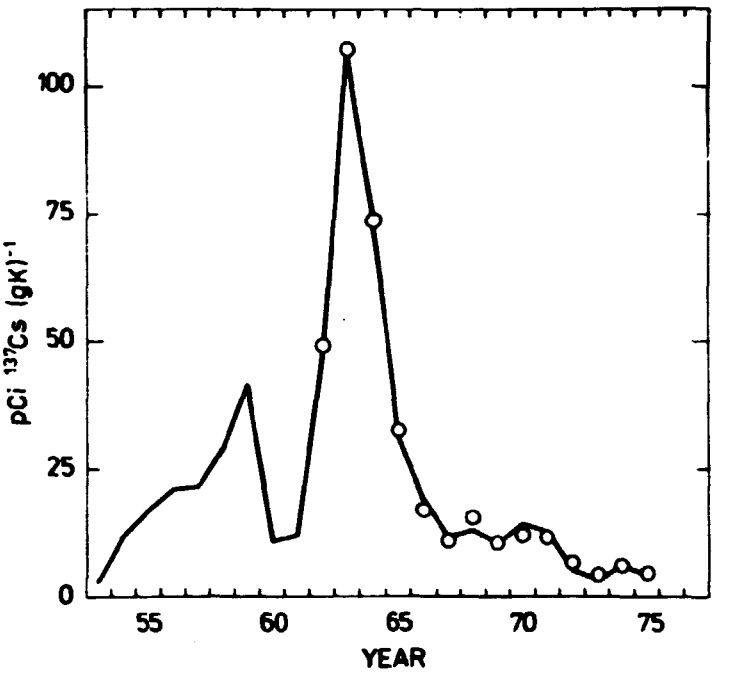

Fig. 3.2.2.3. Predicted (the curve calculated from Table C.3.2.2. No. 1) and observed pCi ^{137}Cs (g K)$^{-1}$ levels in dried milk from Jutland; r = 0.9990***.

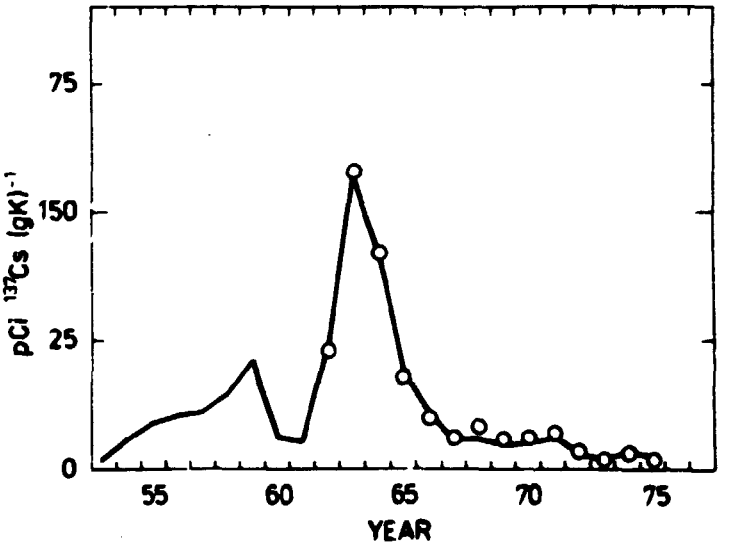

Fig. 3.2.2.4. Predicted (the curve calculated from Table C.3.2.2. No. 3) and observed pCi ^{137}Cs (g K)$^{-1}$ levels in dried milk from the Islands; r = 0.9976***.

The prediction models for milk and table 2.4.2. permit an estimation of the transfer factors from cattle fodder to cows' milk. If the six winter months are assumed represented by winter plan 18 and the summer month by plan S in table 2.4.2., the infinite time-integrated mean intakes of an average cow from a deposition of 1 mCi km^{-2} of the nuclide in question becomes 0.82 µCi ^{90}Sr and 0.29 µCi ^{137}Cs. The infinite time-integrated secretion with milk of the average cow becomes $3.25 \cdot 1.2 \cdot 11 \cdot 365 \cdot 10^{-6} = 0.016$ µCi ^{90}Sr per mCi ^{90}Sr km^{-2} and $3.43 \cdot 1.6 \cdot 11 \cdot 365 \cdot 10^{-6} = 0.022$ µCi ^{137}Cs per mCi ^{137}Cs km^{-2}. (Milk contains 1.2 g Ca and 1.6 g K per litre and a cow produces on the average 11 l milk day^{-1}). Hence, 1.9% of the ^{90}Sr in the fodder was secreted in the milk, or 0.17% per litre, and 7.6% of the ^{137}Cs intake, or 0.7% per litre. The secretions are compatible with the observations of other authors. COX et al. (Co60) thus observed in a field study of a dairy herd that 1.2% of the total ingested ^{90}Sr (and stable Sr) was secreted into the milk. In an experimental study, COMAR et al. (Co61b) found that 0.08% of the daily radiostrontium dose was secreted per liter of milk. In both studies the daily calcium intakes were approx. 100 g per cow. From flow and balance considerations for stable Sr in the cow, COMAR (Co66a) has estimated that 4% of the daily Sr intake would appear in the milk if the daily calcium intake was 50 g; in the case of 100 g Ca d^{-1}, the secretion would be 2% provided that the observed ratio: $\frac{(Sr/Ca)milk}{(Sr/Ca)fodder}$ was constant ($\sim$ 0.1).

In the case of ^{137}Cs, the percentage of cæsium ingested daily that may be found in milk under conditions of prolonged exposure ranged from about 6 to 12 per cent for cows (Fr66).

In the food chain, the so-called observed ratio, OR, between a radionuclide and its congener in step (j) and the preceding (i) is often considered (Co56). In the case of ^{90}Sr:

$$OR_{j/i} = \frac{pCi\ ^{90}Sr\ (g\ Ca)^{-1}\ (j)}{pCi\ ^{90}Sr\ (g\ Ca)^{-1}\ (i)} .$$

The steps (j) and (i) are denoted sample and precursor, respectively.

The OR values are supposed to be constant within a limited range of calcium concentrations. Hence, if the calcium concentration in the precursor is increased by a certain factor and the amount of ^{90}Sr is kept constant, we may expect a reduction in the pCi ^{90}Sr (g Ca)$^{-1}$ ratio by the same factor in the sample. If the precursor is cattle feed and cows' milk is the sample, the observed ratio is 0.1 (range 0.08-0.16) (Co66a) according to numerous investigations. The average daily Ca intake per cow fed Danish fodder of the above-mentioned composition was approx. 60 g (Br69), and the daily secretion of Ca with the daily milk production of the cow was 13 g. Hence, the $OR_{milk/fodder} = \frac{0.016/13}{0.82/60} = 0.089$. Besides the 60 g Ca in the fodder, mineral calcium is often added to the fodder during the winter; 50 g dicalcium-phosphate is thus recommended in winter plan 18 (table 2.4.2.), this increased the annual OR value to 0.10. If the total daily calcium intake the year round was 100 g, the OR became 0.15. The present radioecological sensitivities for ^{90}Sr in fodder and milk thus yielded $OR_{milk/fodder}$ values in agreement with other observations.

3.2.3. Short-lived nuclides in Danish milk

During periods with atmospheric nuclear test-explosions, short-lived nuclides, such as ^{89}Sr ($t_{1/2}$: 50.5 days) and ^{131}I ($t_{1/2}$ = 8.04 days), may appear in milk if the cows are grazing when the fresh fallout is received. During 1961-1963 (RRD61, RRD62, RRD63), in connection with the intensive test series in 1961-1962, these two short-lived nuclides were studied in Danish milk.

Strontium-89 was measured from October 1961 to December 1963. The time-integrated level became 116 pCi ^{89}Sr (g Ca)$^{-1}$ y in Danish dried milk. This level originated from a total deposition of 269 mCi ^{89}Sr km^{-2} (RRD59-76). Hence, the transfer factor became 0.43 pCi ^{89}Sr (g Ca)$^{-1}$ y per mCi ^{89}Sr km^{-2}, or 13% of the corresponding factor for ^{90}Sr. From ^{89}Sr milk measurements in 63 cities in the United States, UNSCEAR (Un77) estimated 0.38 pCi ^{89}Sr (g Ca)$^{-1}$ y per mCi ^{89}Sr km^{-2}. Due to the short half-life of ^{89}Sr, milk was significantly less sensitive to contamination by this nuclide than by the long-lived ^{90}Sr.

Indirect contamination from root uptake thus played no role for ^{89}Sr in milk, and the stored fodder used during the winter lost its content of ^{89}Sr more rapidly than it did ^{90}Sr.

As ^{89}Sr in milk only depended on direct contamination, just like ^{137}Cs, the two nuclides could obviously be compared. An anova of $^{137}Cs/^{89}Sr$ in dried milk showed a probably significant variation between locations, besides the highly significant variation between months. The $^{137}Cs/^{89}Sr$ ratio in milk from Lolland-Falster was thus probably less than in milk from West Jutland, which agrees with the observations for the $^{137}Cs/^{90}Sr$ ratios (3.2.1). As there was no significant difference between the $^{89}Sr/^{90}Sr$ ratios in milk from West Jutland and from Lolland-Falster, the observations suggested that the fodder in West Jutland was relatively more sensitive to direct contamination by ^{137}Cs than by radiostrontium, than the fodder in Lolland-Falster. This observation did not preclude that the fodder also differed with respect to indirect contamination by ^{90}Sr. In nuclear weapons the ratio between ^{89}Sr and ^{90}Sr at detonation is 185 (Un77), and the mean ratio between the deposited activities in Denmark was measured to be 6.3 from the 1961-1962 test explosions (RRD59-76). Hence, the infinite time-integrated ^{89}Sr milk levels from atmospheric nuclear explosions in the Northern hemisphere in 1961-1962 were approximately 80% of the corresponding ^{90}Sr level in milk from these explosions.

Iodine-131 in Danish milk has been measured in three periods: September-November 1961 (RRD61), September-November 1962 (RRD62) and in October 1976 (RRD76). In all periods samples were obtained from farms near Risø, and in 1961 and 1962 they were also collected countrywide. The 1961 study showed that samples from Jutland contained approx. 20% more ^{131}I than samples simultaneously collected from the Islands. The time-integrated levels in Danish milk in 1961 were estimated at 2-3 pCi ^{131}I l^{-1} y, in 1962 at 5-6 pCi ^{131}I l^{-1} y and in 1976 at 0.5-1 pCi ^{131}I l^{-1} y. In 1962 and 1976 the deposition of ^{131}I was measured; from this were estimated the transfer factors from deposit to milk. In 1962 the factor was 0.7 pCi l^{-1} y per mCi ^{131}I km^{-2} and in 1976 0.5 pCi l^{-1} y per mCi ^{131}I km^{-2}. These

estimates were based upon observations made in the last months of the summer grazing season and the first of the winter stall season. The factors were thus lower than expected for midsummer, but defintely higher than those for the winter, and they may correspond to the annual mean situation. As compared with transfer factors from other countries (Un72) with a mean value of 2.2. pCi $y\ l^{-1}$ per mCi km^{-2} (range: 0.6-4.1), the Danish factors were low. In 1962 the concentrations in grass and milk were compared in samples collected from the state experimental farms in September (RRD62). The mean ratio pCi $^{131}I\ l^{-1}$ milk/ pCi $^{131}I\ kg^{-1}$ grass (dry matter) (20% dry matter) was 0.0065 (1 SE 0.001). From simultaneous measurements of $^{89}Sr/^{90}Sr$ in milk and grass, it was estimated that 67% of the cattle fodder was freshly contaminated, and it was assumed that this part of the fodder consisted of grass. Hence, for a pure grass diet, the activity ratio between milk and grass became 0.01. This estimate was consistent with the majority of values found in field studies (Ga66). The relatively low sensitivity of Danish milk to contamination by ^{131}I was thus due to the high productivity of the grazing areas (cf.2.4.1.) combined with the relatively large amounts of supplementary fodder, and to the fact that Danish cows are stalled for half of the year, which implies that short-lived nuclides disappear from the stored fodder.

3.3. Faroese milk

Er det en Høj sprøjte den mindste af Drengene og pegede på Elverhøj. "Det kalde vi oppe i Norge et Hul!
ELVERHØJ

"Do you call that thing a hill?" asked the youngest of them, pointing to the Elf Hill, "Up in Norway we should call it a cave."
THE HILL OF THE ELVES

A combination of high rainfall and specific agricultural conditions may result in enhanced radionuclide concentrations in milk. Already by the end of the fifties, such sites were identified in the United Kingdom (Ag60) and in Norway (Hv61). Later, similar regions (Un69) have been found in Florida, New Zealand, the Ukraine, Jamaica and in the Faroe Islands. The Norwegian study showed that high ^{137}Cs milk levels might occur even for relatively low fallout rates. Agricultural conditions may thus be more important than rainfall. In the Faroes, precipitation is high ($\sim$ 1500 mm yr^{-1}), farming is

extensive, and the mineral content of the soil is relatively low while the organic matter content is high; an enhanced content of radioactivity in Faroese milk was thus to be presumed.

3.3.1. Variation with time and location

The variation of the annual ^{90}Sr and ^{137}Cs concentrations in Faroese milk was highly significant (tables B.3.3.1. and

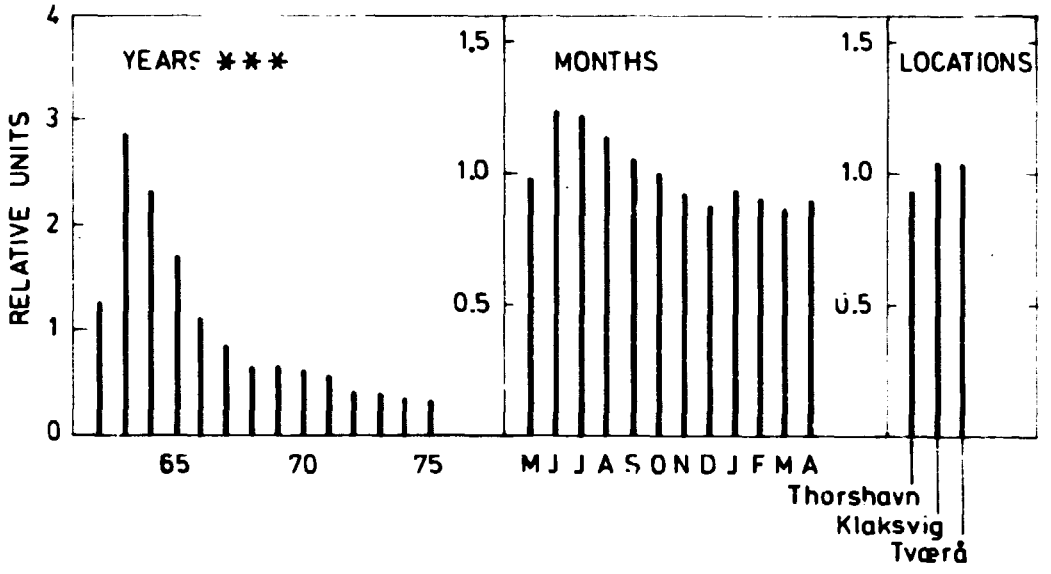

Fig. 3.3.1.1. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in fresh Faroese milk collected weekly at 3 locations (Fig. A.1.1.3.2.) from May 1962 to April 1976 (cf. Table A.1.3.3.). The bars indicate the levels relative to the grand mean 57 pCi (g Ca)$^{-1}$ (= 1 at the relative scales). The years were "milk years".

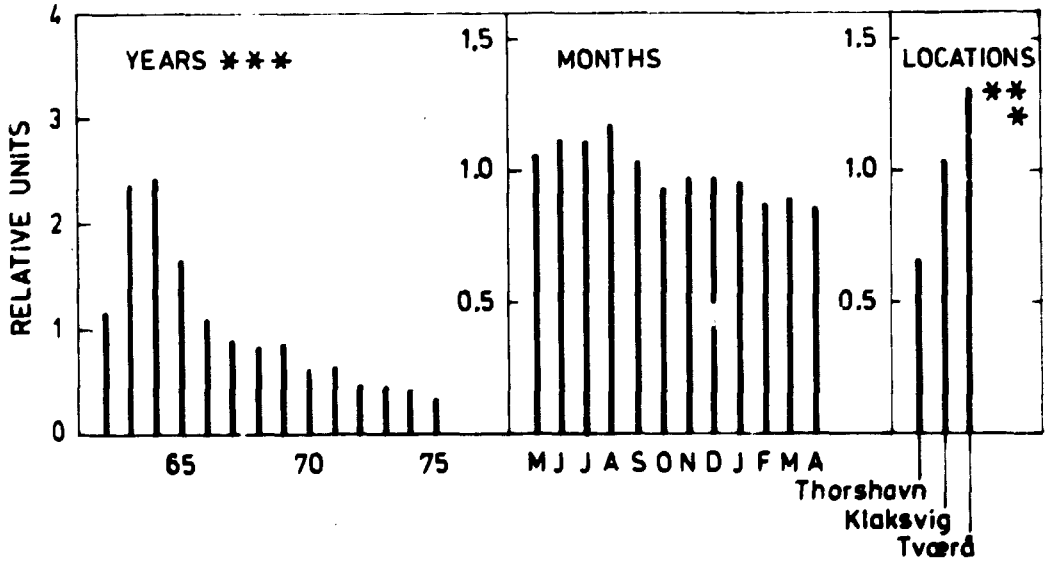

Fig. 3.3.1.2. The variation of pCi ^{137}Cs (g K)$^{-1}$ in fresh Faroese milk collected weekly at 3 locations (Fig. A.1.1.3.2.) from May 1962 to April 1976 (cf. Table A.1.3.3.). The bars indicate the levels relative to the grand mean 329 pCi (g K)$^{-1}$ (= 1 at the relative scales). The years were "milk years".

B.3.3.2.). The annual maximum ^{90}Sr levels in 1963-1964 were approx. 7 times higher than the concentrations found in 1973-1975, and the ^{137}Cs levels were 5-6 times higher (figs. 3.3.1.1. and 3.3.1.2.). The variability (table B.3.3.3.) of the annual ^{90}Sr levels in milk levels was 0.73, and that of the ^{137}Cs concentrations was 0.67; the variability of the two nuclides was not significantly different. This was contrary to the observations concerning Danish milk (3.2.1.), where the annual variability was 1.45 for ^{137}Cs and 0.70 for ^{90}Sr.

The variation among the monthly Faroese milk concentrations was not significant for ^{137}Cs but was significant for ^{90}Sr in some years. The ^{90}Sr levels in the summer months were thus generally 20-25% higher than those in the rest of the year. The variabilities among months were 0.09 and 0.12 for ^{137}Cs and ^{90}Sr, respectively. Once again there is an evident difference compared to the Danish time pattern, where the ^{137}Cs milk levels during the summer months were significantly higher than during the winter ($CV_{p\ month} = 0.29$), but where, on the other hand, the ^{90}Sr concentrations showed no marked variations throughout the year ($CV_{p\ month} = 0.04$).

As regards the local variations, there was only little difference between the ^{90}Sr concentrations in milk from the 3 locations ($CV_{p\ location} = 0.05$). The ^{137}Cs levels, however, differed significantly ($CV_{p\ location} = 0.29$). Milk from Tværå thus contained nearly 80% more ^{137}Cs than milk from Thorshavn. Compared with Danish milk, the ^{90}Sr concentrations in Faroese milk have been 4-6 times those in Danish, while the ^{137}Cs levels in Faroese milk have been 8 to 44 times higher than the Danish levels, with the highest quotients in the later years. It is remarkable that the local variability for ^{137}Cs in Faroese milk has shown an increasing tendency with time (table B.3.3.3.), and that the variability among years was lower for Tværå than for Thorshavn and Klaksvig. The marked difference between the time variation of ^{137}Cs in Faroese and in Danish milk was primarily ascribed to the indirect contamination of Faroese cattle feed with ^{137}Cs from root uptake (cf.2.4.1.). Consequently, the Faroese ^{137}Cs levels in milk were influenced not only by the fallout rate, as the Danish milk, but also by the accumu-

lated fallout, and as the fallout rate decreased the indirect contamination became relatively more important, which caused an increasing local variability. The lower variability among years observed for ^{137}Cs in Tværå milk suggests that the levels in milk from this location were more influenced by indirect contamination of the cattle feed than was the case in the two other localities.

3.3.2. Relations and prediction models

As the local variation of the ^{90}Sr concentrations in Faroese milk was modest, the prediction models for ^{90}Sr in Faroese milk were calculated from the mean of the 3 locations (table C.3.3.1). The radioecological sensitivity of Faroese milk (8.6 pCi ^{90}Sr (g Ca)$^{-1}$ y per mCi ^{90}Sr km^{-2}) was 2.6 times that of Danish milk for ^{90}Sr (table C.3.2.1). As the ^{90}Sr fallout rate was 2.08 times higher in the Faroes than in Denmark, the infinite time-integrated ^{90}Sr concentration in Faroese milk was 5.5 times that in Danish milk.

The radioecological sensitivity of Faroese milk to ^{137}Cs contamination was approx. twice as high in Tværå than in Thorshavn and Klaksvig. The mean sensitivity for Faroese milk (table C.3.3.2) was 35 pCi ^{137}Cs (g K)$^{-1}$ y per mCi ^{137}Cs km^{-2}, or 59 pCi ^{137}Cs l^{-1} y per mCi ^{137}Cs km^{-2}. This was a 10 times higher sensitivity than that observed for Danish milk (table C.3.2.2.). The infinite time-integrated ^{137}Cs level in Faroese milk was thus approx. 20 times higher than that in Danish milk.

According to the Faroese Agricultural Adviser (Wa74), 38% of the feed units consumed by Faroese cows were imported concentrates. By the middle of the seventies, Faroese cows received 8 feed units per day and they produced approx. 2500 kg milk yr^{-1} (3.7% fat). In analogy with the secretion estimates for Danish milk (cf.3.2.2.), the secretions of ^{90}Sr and ^{137}Cs in Faroese milk were estimated at 0.1-0.3% and 0.4-0.7% per l of the daily intakes, respectively. This was compatible with the Danish estimates. The estimates for Faroese milk assumed that the Faroese fodder consisted of 38% imported Danish barley and 62%

Faroese grass (Wa74). The activity contributions from the concentrates (the barley grain) were negligible as compared with those from the Faroese grass, which were estimated from the radioecological sensitivities shown in 2.4.1.

At the beginning of the sixties, the annual milk production in the Faroes was 1200 kg per cow and each cow received approximately 6 feed units (Wa74). The increase in the consumption of imported concentrates since then may have contributed to a reduction of the radioecological sensitivity of Faroese milk.

3.4. Meat from terrestrial animals

Det var et godt Faar, godt i Stand og godt med Uld. "Det gad jeg nok eje! tænkte Bonden. "Det vilde ikke komme til at savne Græsning på vor Grøftekant.

HVAD FATTER GJØR, DET ER ALTID DET RIGTIGE

"I could do with that sheep, I could" thought the farmer. "It would find plenty of grazing at the side of our ditch.

DAD'S ALWAYS RIGHT

Besides milk and cereal grain, meat is one of the important ^{137}Cs donors in the western diet. In certain arctic population groups, where reindeer or caribou are the main source of meat, the ^{137}Cs whole-body concentrations have been orders of magnitude higher than in the general population of the northern hemisphere (Li61, Han73). In the food chains, ^{137}Cs is often concentrated compared to the congener potassium. It has thus been observed that meat displays a $^{137}Cs/K$ quotient that is approx. 3 times that of the fodder. Meat does not contain substantial amounts of ^{90}Sr and is thus relatively unimportant as a ^{90}Sr donor in the total diet. Animal bones are, however, useful indicator samples for ^{90}Sr contamination of the terrestrial environment.

3.4.1. Danish beef and veal

Anovas of pCi ^{137}Cs $(kg)^{-1}$ in beef and veal collected from $June_{(i)}$ to $March_{(i+1)}$, i.e. within the "milk year" (3.2.1.), showed significant variation among years and among sampling months. The maximum levels occurred in 1963 (fig.3.4.1.), and within the year September and June showed the highest levels,

in agreement with the observations for milk. The ^{137}Cs concentrations in beef and veal did not differ significantly. The ^{90}Sr levels in beef were approx. 40 times lower than the ^{137}Cs levels. In general, the ^{90}Sr levels decayed more slowly than the ^{137}Cs levels, which is also in agreement with the milk observations. The variabilities of ^{137}Cs for beef and veal among years were 1.16 and among months: 0.24 (table B.3.4.1.), while for ^{90}Sr they were 0.60 and 0.17, respectively. Table B.3.2.3. shows that the variabilities among years for milk were similar to those for beef and veal.

The ^{137}Cs contents of beef and veal were thus correlated to that of milk, the concentration of the meat was 4-5 times that of the milk; similar factors have been observed in Sweden (Li65). The transfer factor for ^{137}Cs to beef from fallout was 27 pCi

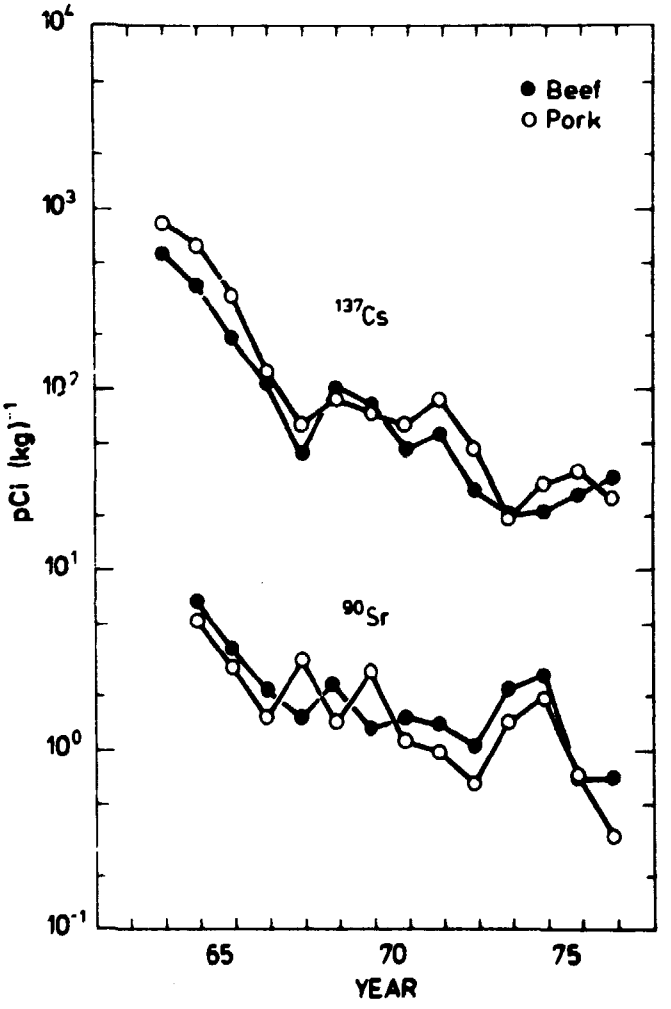

Fig. 3.4.1. Annual means of ^{90}Sr and ^{137}Cs in Danish beef and pork collected from June 1963 - March 1976 in Copenhagen (cf. Table A.1.3.4.1.). The annual means were based on "milk years" (cf. the text).

$^{137}Cs\ kg^{-1}$ y per mCi $^{137}Cs\ km^{-2}$ (table C.3.4.2., nos. 1 and 2), and for ^{90}Sr it was 1.4 pCi $^{90}Sr\ kg^{-1}$ y per mCi $^{90}Sr\ km^{-2}$ (table C.3.4.1., nos. 1 and 2). Beef was thus approx. 20 times more sensitive to contamination by ^{137}Cs than by ^{90}Sr. Beef was approx. 5 times more sensitive to radioactive contamination by ^{137}Cs than was milk, and approx. 3 times less sensitive than milk with respect to contamination by ^{90}Sr (pCi kg^{-1} figures). The total infinite time-exposure integrals in beef from the deposition of 73 mCi $^{90}Sr\ km^{-2}$ and 117 mCi $^{137}Cs\ km^{-2}$ became 3.2 nCi $^{137}Cs\ kg^{-1}$ · y and 100 pCi $^{90}Sr\ kg^{-1}$ · y.

3.4.2. Danish pork

Approximately two-thirds of the meat consumed in Denmark consists of pork, while the remainder is mainly beef and veal. (RRD62). The ^{137}Cs and ^{90}Sr concentrations in pork resemble those in beef (fig.3.4.1.), although in pork the ^{137}Cs levels have been generally higher and the ^{90}Sr concentrations lower. Pork is thus an important contributor to the ^{137}Cs content of the Danish diet. The variability of ^{137}Cs among years was 1.57, i.e. higher than for beef, but among months it was only 0.09, i.e., lower than for beef (table B.3.4.1.). Besides metabolic differences between pigs and cattle, the fodder of the two species may have influenced the radionuclide concentrations of the meat. The fodder of pigs mainly consists of barley, crushed soya and skimmed milk, while grass, grain and beets are the main constituents of cattle feed (Da57-77). Moreover, while cattle feed shows a pronounced seasonal variation due to summer grazing, this is not the case to the same extent for pig fodder.

The transfer factors from fallout to pork were 34 pCi $^{137}Cs\ kg^{-1}$ y per mCi $^{137}Cs\ km^{-2}$ (table C.3.4.2., nos. 3 and 4) and 0.9 pCi $^{90}Sr\ kg^{-1}$ y per mCi $^{90}Sr\ km^{-2}$ (table C.3.4.1., nos. 3 and 4). The factors thus resembled those estimated for beef; but pork was apparently slightly more sensitive to radioactive contamination by ^{137}Cs than was beef. The total infinite time-exposure integrals in Danish pork from fallout were 4.0 nCi $^{137}Cs\ kg^{-1}$ y and 66 pCi $^{90}Sr\ kg^{-1}$ y.

3.4.3. Faroese and Greenlandic mutton

Since the beginning of the sixties ^{137}Cs and ^{90}Sr have been determined in samples of Faroese and Greenlandic sheep. Both mutton and bone have been measured; bone was included in the studies because it is a more sensitive indicator of ^{90}Sr uptake than meat. In the Faroes some 40 000 (Ri72) and in Greenland approx. 20 000 sheep and lambs (Da70) are slaughtered annually. Sheep graze nearly the whole year round and get little - if any - additional fodder. As they often feed on very sparse vegetation, they collect nuclear debris deposited over relatively large areas, and because of the often low productivity of the fields where sheep normally graze the concentration of radionuclides in their fodder is relatively high (Cham70).

The anovas showed that the ^{137}Cs concentration (pCi ^{137}Cs (g K)$^{-1}$) in Faroese mutton was 1.8 times that of Greenland mutton (fig.3.4.3.1.) and the ^{90}Sr content of sheep bone was 2.7 times higher in the Faroes than in Greenland (fig.3.4.3.2). These factors may be compared with the approximately 2.8 times higher

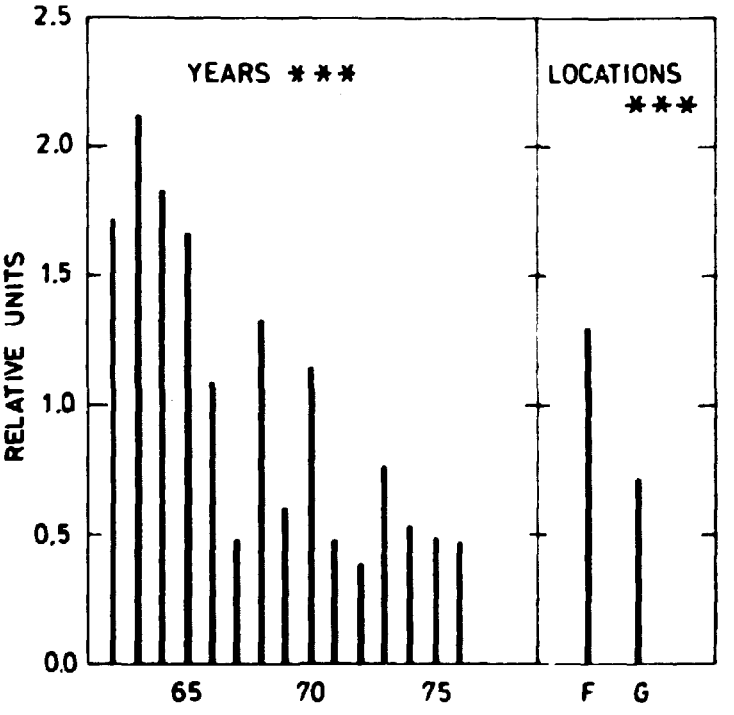

Fig. 3.4.3.1. The variation of nCi ^{137}Cs (g K)$^{-1}$ in mutton collected in 1962-1976 in the Faroes and in Greenland (Table A.1.3.4.2.). The bars show the levels relative to the grand mean 0.66 nCi (g K)$^{-1}$ (= 1 at the relative scale).

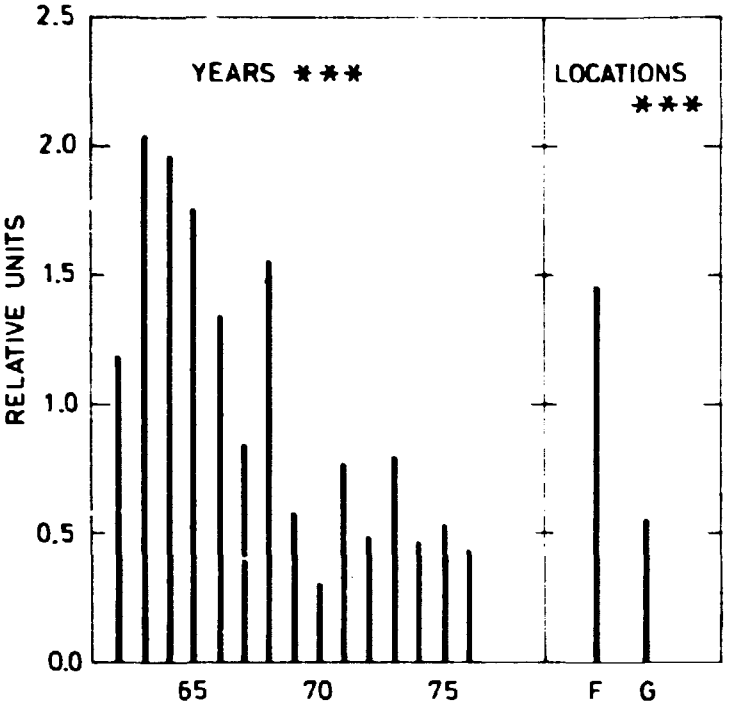

Fig. 3.4.3.2. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in sheep bone collected in 1962-1976 in the Faroes and in Greenland (Table A.1.3.4.2.). The bars show the levels relative to the grand mean 200 pCi (g Ca)$^{-1}$ (= 1 at the relative scale).

fallout rate in the Faroes than in West Greenland (Godthåb). The variabilities of ^{137}Cs among years were 0.79 in Faroese and 0.99 in Greenland mutton, i.e. not significantly different (table B.3.4.1.).

As some of the samples received from the Faroes consisted of dried meat it was expedient to use the quotients $^{137}Cs/K$ and $^{90}Sr/Ca$ in the comparisons between Faroese and Greenlandic mutton instead of pCi kg^{-1}. The radioecological sensitivities (tables C.3.4.1. and C.3.4.2.) of Faroese mutton were 79 pCi ^{137}Cs (g K)$^{-1}$ y ($\sim$ 205 pCi ^{137}Cs kg^{-1} y) per mCi ^{137}Cs km^{-2} and 31 pCi ^{90}Sr (g Ca)$^{-1}$ y ($\sim$ 3.1 pCi ^{90}Sr kg^{-1} y) per mCi ^{90}Sr km^{-2}. The corresponding values for Greenland mutton were 135 ($\sim$ 355) and 53 ($\sim$ 5.3), respectively. Although the radioecological sensitivities were apparently higher in Greenland than in the Faroes, the difference may not be significant because the Greenland data were incomplete. Table C.3.4.3. presents an estimate of the radioecological sensitivities of ^{90}Sr in sheep bone. It appears that in this case the Faroes and Greenland showed the same transfer factors, as also expected

from the above-mentioned anova. The total infinite exposure integrals of Faroese mutton from fallout from nuclear weapons testing were 50 nCi ^{137}Cs kg^{-1} y and 0.5 nCi ^{90}Sr kg^{-1} y, and for Greenland mutton the levels were 31 nCi ^{137}Cs kg^{-1} y and 0.3 nCi ^{90}Sr kg^{-1} y.

3.4.4. Greenland reindeer and musk ox

Since 1961 samples of reindeer meat and bone have been obtained from Greenland. The main part of the material was collected between Godthåb in the south and Egedesminde in the north during the fall (August-October) and in late winter (February-March). The samples consisted of wild reindeer (Rangifer tarandus groenlandicus) as well as domestic animals. Reindeer were also purchased from the Royal Greenland Trading Company; the exact geographic location of these samples was unknown, but they were allocated to the south-west coast in general. There are approximately 40 000 reindeer in Greenland and of these approximately 3 000 are domestic animals concentrated along the Godthåb Fjord. In 1967 approximately 7 000 animals were slaughtered (Vi71).

The anova showed no significant local variation in the ^{137}Cs and ^{90}Sr concentrations of the samples. This was compatible with similar observations for grass (2.4.1) and for lichen (2.6.1.) from the south-west coast of Greenland. The time variation was, however, significant (figs. 3.4.4.1 and 3.4.4.2). From the first half of the sixties to the first half of the seventies, both the ^{90}Sr and the ^{137}Cs levels in the meat decreased by an order of magnitude. The variabilities among years were 1.6 for ^{137}Cs and 1.1 for ^{90}Sr, and thus they were higher than those observed for the fodder of reindeer (cf. tables B.2.4.1 and B.2.6.1). The higher variabilities in the reindeer were due to relatively high activity samples from 1967 and 1968. Individual extremes, which carried much weight in the limited number of samples, may thus have produced this apparent difference among the variabilities of fodder and reindeer. In agreement with the studies of HANSON (Ha73) in Alaska and of MIETTINEN (Mie63), PERSSON (Per70) and SWEDJEMARK (Sw74) in Lapland, the activity levels in Greenland reindeer

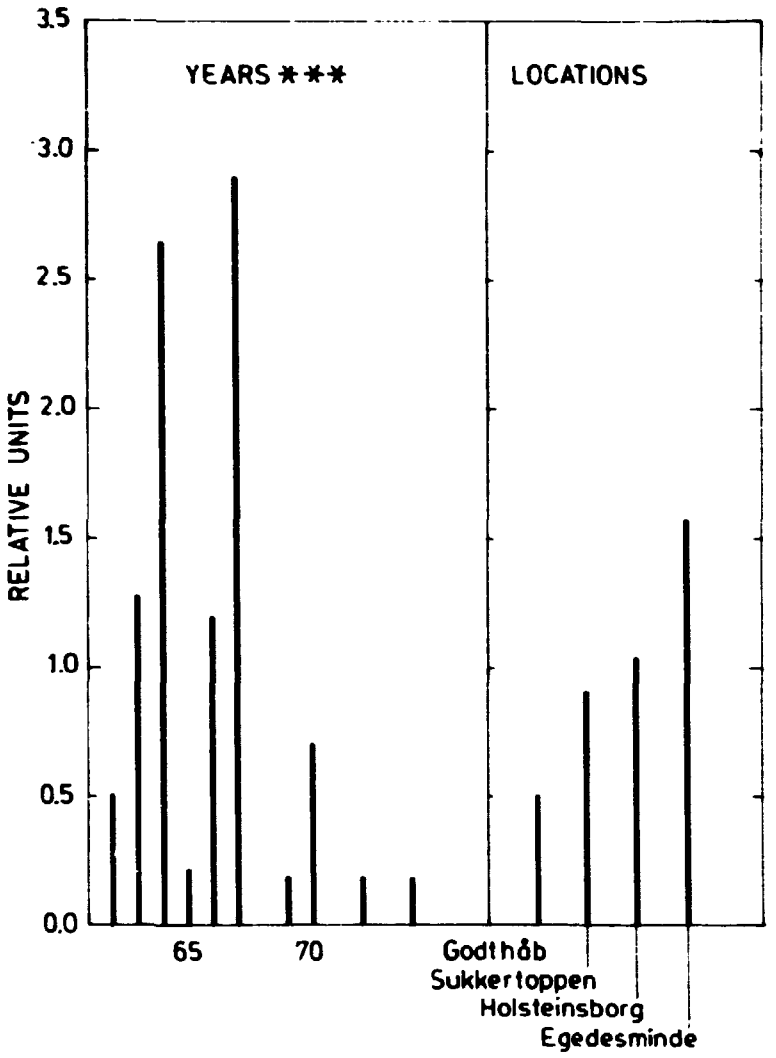

Fig. 3.4.4.1. The variation of nCi ^{137}Cs kg^{-1} in reindeer meat collected in 1962-1974 in West Greenland (Table A.1.3.4.2.). The bars show the concentrations relative to the grand mean 5.8 nCi kg^{-1} (= 1 at the relative scale).

samples also showed seasonal variations. The ^{137}Cs levels in reindeer meat from the late winter and the spring were thus approx. three times higher than in meat from the fall, while the ^{90}Sr concentrations were twice as high. As shown in the Alaskan and Lapland studies, this agrees with the feeding habits of reindeer, which generally eat lichen during the winter but in summer generally feed on vegetation with a lower content of radioactivity such as grass, sedges, herbs, mushrooms and leaves from bushes. The ^{137}Cs levels in Greenland were significantly lower than the corresponding concentrations in meat from Alaska and Lapland, which contained respectively two and three times more ^{137}Cs; however, in the far north of the USSR local variations of more than a factor of three are not

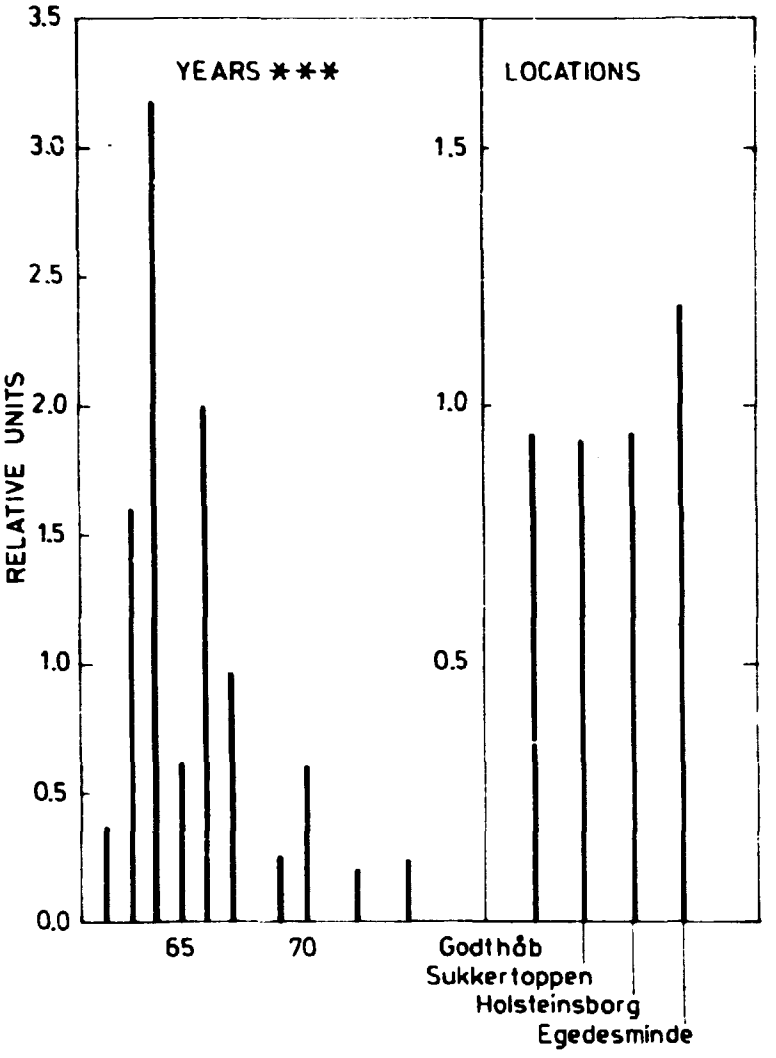

Fig. 3.4.4.2. The variation of pCi $^{90}Sr\ kg^{-1}$ in reindeer meat collected in 1962-1974 in West Greenland (Table A.1.3.4.2.). The bars show the concentrations relative to the grand mean 45 pCi kg^{-1} (= 1 at the relative scale).

unusual (Ne66). As regards ^{90}Sr, the Greenlandic levels in meat as well as in bone were close to the Swedish ones, but significantly higher than the Alaskan values, which were one third to one half of the Greenland and Lapland concentrations. As the arctic regions showed only minor differences with respect to the ^{137}Cs and ^{90}Sr concentrations in lichen (2.6), the observations suggested that the local differences in the reindeer levels depended on factors other than the radionuclide concentration in lichens; feeding habits may thus show local variations. NEVSTRUEVA et al. (Ne66) draw a similar conclusion from their studies in the arctic USSR.

The prediction models for reindeer (cf.D.3.4.4) showed that the transfer factors for meat were 1.5 nCi $^{137}Cs\ kg^{-1}$ y per mCi $^{137}Cs\ km^{-2}$ (table C.3.4.2., nos. 9 and 10) and 15 pCi $^{90}Sr\ kg^{-1}$ y per mCi $^{90}Sr\ km^{-2}$ (table C.3.4.1., nos. 8 and 9), or 120 pCi $^{90}Sr\ (g\ Ca)^{-1}$ y per mCi $^{90}Sr\ km^{-2}$ and in bone 100 pCi $^{90}Sr\ (g\ Ca)^{-1}$ y per mCi $^{90}Sr\ km^{-2}$ (table C.3.4.3., no. 5). Prediction models for the meat collected in the late winter showed transfer factors approx. three and two times higher for ^{137}Cs and ^{90}Sr, respectively, than for meat collected in the fall. The radioecological sensitivity of reindeer to ^{137}Cs contamination was 4-5 times higher than that of Greenland sheep (3.4.3.), which is compatible with the observations of HANSON (Han73), who found 4-10 times more ^{137}Cs in caribou flesh than in Dall sheep.

The total infinite time-integral in Greenland reindeer meat from fallout became 132 nCi $^{137}Cs\ kg^{-1}$ y and 0.8 nCi $^{90}Sr\ kg^{-1}$ y.

The musk ox (Ovibos moschatus) lives in north-east Greenland. By the end of the sixties there were 6 000-12 000 animals, according to VIBE (Vi71). The musk ox is most frequently found in the Scoresbysund Fjord district, at Kejser Franz Joseph Fjord, at Danmarkshavn and in Peary Land. Samples of musk ox were obtained from north-east Greenland during 1964-1973. The present data are the only information available on ^{90}Sr and ^{137}Cs levels in musk ox.

The median concentrations in musk ox meat during 1964-1973 were 140 pCi $^{137}Cs\ kg^{-1}$ and 8 pCi $^{90}Sr\ kg^{-1}$, and in bone 60 pCi $^{90}Sr\ (g\ Ca)^{-1}$. The levels have shown a decreasing tendency similar to that observed for reindeer samples. The variability among years in musk ox was 0.8 for both ^{137}Cs and ^{90}Sr, i.e. close to that observed for sheep. It was evident that the ^{137}Cs and ^{90}Sr levels in musk ox were significantly lower than those in reindeer. The potassium content of the musk ox meat samples was approx. 75% of that of the reindeer samples. This feature, together with the different feeding habits of the animals, contributed to the relatively lower ^{137}Cs concentrations in the flesh of the musk ox.

The limited number of data and the great local variations in precipitation, and thus in the fallout levels, between the Scoresbysund Fjord districts and Danmarkshavn (the areas from where the samples were obtained) make the calculation of prediction models problematic. The precipitation at Scoresbysund and on the south-west coast of Greenland is 4-5 times higher than that in Danmarkshavn; but the low precipitation at Danmarkshavn may yield an underestimate of the fallout at this station if dry fallout makes significant contributions (cf. also D.3.4.4.). The radionuclide levels in moss and lichen from Danmarkshavn (RRG62-76) indicated that the fallout levels may have been a little lower than in other parts of Greenland, but certainly not 4-5 times lower. As the best available estimate of fallout in North-East Greenland, measurements from Kap Tobin at Scoresbysund were therefore applied. This may be an overestimate of the actual mean fallout for North-East Greenland, and the transfer factors calculated may thus be too low. The radioecological sensitivities of musk ox meat were 0.08 nCi $^{137}Cs\ kg^{-1}$ y per mCi $^{137}Cs\ km^{-2}$ and 4 pCi $^{90}Sr\ kg^{-1}$ y per mCi $^{90}Sr\ km^{-2}$ (tables C.3.4.1. and C.3.4.2.). The infinite time-integrals until 1975, i.e. from a total deposit of 55 mCi $^{137}Cs\ km^{-2}$ and 34 mCi $^{90}Sr\ km^{-2}$ from fallout became 4.4 nCi $^{137}Cs\ kg^{-1}$ y and 0.14 nCi $^{90}Sr\ kg^{-1}$.

The radioecological sensitivities of musk ox meat to contamination by ^{90}Sr and ^{137}Cs were less than for reindeer meat, but in the case of ^{90}Sr the difference may not be significant because of the uncertainty of the fallout data used for musk ox. The difference in radioecological sensitivity between the two animals was ascribed to the different feeding habits. While lichen is important to reindeer, musk oxen prefer grass, sedges, leaves and twigs of small bushes, and during the winter they eat arctic willow, but if starving they will consume anything.

3.5. Fish

Fisken var blevet fanget, kørt paa Torvet, solgt og kommen op i Kjøkkenet hvor Pigen skar den op med en stor Kniv.
DEN STANDHAFTIGE TINSOLDAT

The fish had been caught, taken to market and sold, and here it was in the kitchen, where the maid cut it open with a big knife.
THE STAUNCH TIN SOLDIER

Fish obtain radioactive substances via two main pathways: through absorption of the surrounding water by their gills and from the ingestion of food, which again directly or indirectly depends upon the radionuclide content of the water. In shallow waters direct contamination from sediments may be a third exposure route (Lo71).

Fish are not a major component of the Danish diet, the annual mean per caput consumption being of the order of 10 kg. However, in the Faroes and Greenland the annual per caput consumptions are an order of magnitude higher. Nearly all fish consumed in the three countries are of marine origin. Fresh water fish is of very little importance, and enhanced ^{137}Cs levels such as those observed especially in fish from oligotrophic lakes in the other Nordic countries (Ca76, Ko66) play no role for the ^{137}Cs intake with the Danish diet.

3.5.1. Variation with time, location and species

The radioactivity content of marine fish is expected to be closely related to the radionuclide concentrations of sea water. A comparison of the variabilities found in tables B.3.5.1. and B.1.5.1. confirms this expectation. The radionuclide levels in fish from the Faroes and Greenland showed a significantly higher variability among years ($\sim$ 0.5) than Danish fish ($\sim$ 0.3), in accordance with the higher variability observed in the Atlantic ocean ($\sim$ 0.5) than in Danish waters ($\sim$ 0.2). The ^{90}Sr and ^{137}Cs concentrations in fish were highest in the first part of the sixties (fig.3.5.1.). An anova showed that the ^{137}Cs concentrations in Faroese cod fish were 7 times lower than in cod from Denmark, which implied that the ^{137}Cs per caput intake from fish was nearly the same in the two countries, because the Faroese per caput fish consumption was 8 times that of the Danish.

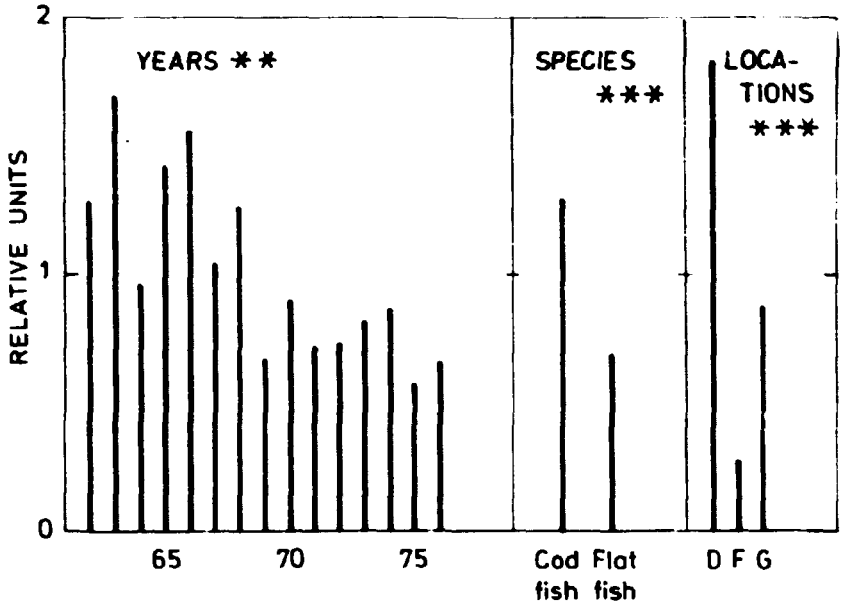

Fig. 3.5.1. The variation of pCi ^{137}Cs kg^{-1} in flesh of cod fish (mostly cod and haddock) and flat fish (mostly plaice and halibut) collected in Denmark, the Faroes and Greenland in 1962-1976 (Table A.1.3.5.). The bars show the concentrations relative to the grand mean 36 pCi kg^{-1} (= 1 at the relative scale).

Cs-137 and ^{90}Sr showed similar variabilities among years; the marked internuclide difference of the variability observed for terrestrial samples was thus absent for fish. The variability among years was lower for fish than for terrestrial animals because of the generally stronger dependence of the marine fauna on the accumulated fallout; the sea acted, so to say, as a buffer and smoothed out short-term variations from the fallout rate.

The ^{90}Sr concentrations in fish flesh were 1-2 orders of magnitude lower than the ^{137}Cs levels, i.e. similar to the ratio observed for terrestrial meat (3.4.1. and 3.4.2.). Fish bone contained ^{90}Sr concentrations that were 3 orders of magnitude higher than those in the flesh, corresponding to the higher content of Ca in bone. The ^{90}Sr concentrations in fish flesh were thus influenced by the presence of even small amounts of bone in the samples (D.3.5.1.).

The anovas showed that cod fish (gadus species) had nearly twice as high concentrations of ^{137}Cs than flat fish (pleuronectes species), which may be a result of the feeding

habits of the two species, the trophic levels of the cod being higher than those of the flat fish. The few samples of euryhaline species (eel, salmon and trout) suggested higher ^{137}Cs as well as ^{90}Sr levels than in other species, in agreement with expectations from the generally higher levels encountered in fresh-water fish. The interspecific variability in Danish fish species was nearly the same for ^{137}Cs and for ^{90}Sr.

3.5.2. Relations and prediction models

The term "concentration factor" (CF) is defined as the ratio of the concentration of the radionuclide in an organism to the concentration directly available from the environment of the organisms under steady-state conditions (Lo71). As organisms may derive radionuclides from various sources, including food, water and sediments, concentration factors are not absolute; they may be altered by biological and environmental factors. POLIKARPOV (Po66) found that the CFs of a given radionuclide within closely related species of marine plants and animals do not differ significantly from each other in different seas and oceans with different salinity (from 35‰ to 17‰). This finding was not fully compatible with the observations of FELDT (Fe66) and BRYAN (Bry66), who found that the CF for ^{90}Sr and ^{137}Cs in marine organisms showed a decrease with increasing salinity.

The prediction models estimated for Danish, Faroese and Greenlandic cod fish (tables C.3.5.1. and C.3.5.2.) and waters (table C.1.5.1.) may be used to estimate the CF in the three areas. As Windscale started to contaminate Danish waters from 1972-1973 (1.5), only samples from before 1972 were included in the Danish models. The transfer factors for ^{137}Cs in cod were 20 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-1} in Denmark, 1.7 in the Faroes and 9 in Greenland. In the case of ^{90}Sr, Danish cod showed a radioecological sensitivity of 0.9 pCi ^{90}Sr $(g\ Ca)^{-1}$ y (or 0.09 pCi ^{90}Sr kg^{-1} y) per mCi ^{90}Sr km^{-2} and Faroese cod fish 0.6 or 0.06, respectively. From these data and the corresponding figures for the radioecological sensitivities of the respective waters (1.5.), the transfer factors from water to fish were estimated at 87, 57 and 39 pCi ^{137}Cs kg^{-1} y per

pCi ^{137}Cs l^{-1} y for Denmark, the Faroes and Greenland, respectively, and for ^{90}Sr at 0.3 and 2 pCi ^{90}Sr kg^{-1} y per pCi ^{90}Sr l^{-1} y for Denmark and the Faroes. The ^{137}Cs figures suggested that the CF was higher in the low salinity Danish waters (16‰) than in the high salinity Faroese and Greenlandic seas (34‰ and 29‰) in agreement with the observations of FELDT (Fe66) and BRYAN (Bry66). However, for ^{90}Sr, the opposite was the case. The prediction models for ^{90}Sr in fish were encumbered with some uncertainty, due partly to the relatively few data available and partly to the varying amounts of bone in the samples. Concentration factors for marine fish in the literature (Ji72) were in the range 5-244 (mean 48) for Cs and 0.1-1.5 (mean 0.43) for Sr.

The radioecological sensitivity of Greenland salmon to ^{137}Cs contamination was similar to that of cod; Danish plaice seemed to be less sensitive to radioactive contamination than Danish cod (tables C.3.5.1. and C.3.5.2.). It was remarkable that the radioecological sensitivity of cod was approx. 75% of that of beef; this relatively modest difference was a result of the slower decrease in the ^{137}Cs levels of the marine fauna than of the terrestrial fauna in Denmark. Although the terrestrial ^{137}Cs levels after global contamination were thus initially higher than the marine levels, the infinite time-integrated levels for environmental samples from the two environments were not much different from each other.

3.6. Various animals

Saa gik det paa Fangst! Harpunen blev sat i Hvalrossens Bryst, saa den dampende Blodstraale stod som et Springvand over Isen.
PARADISETS HAVE

After that came the fishing. The harpoon was plunged in the heart of the walrus, so that the steaming blood spirted up like a fountain over the ice.
THE GARDEN OF EDEN

3.6.1. Sea mammals

Seal and whale are important constituents of the Faroese and Greenlandic diets. In the Faroes, 25% of the "meat and egg" consumed was whale (RRF62), while in Greenland seal and whale contributed 65% (RRG62). The sampling of seal and whale (Piked Whale and Grindhval) from Greenland and the Faroes has been irregular; most samples were received during the first half of the sixties.

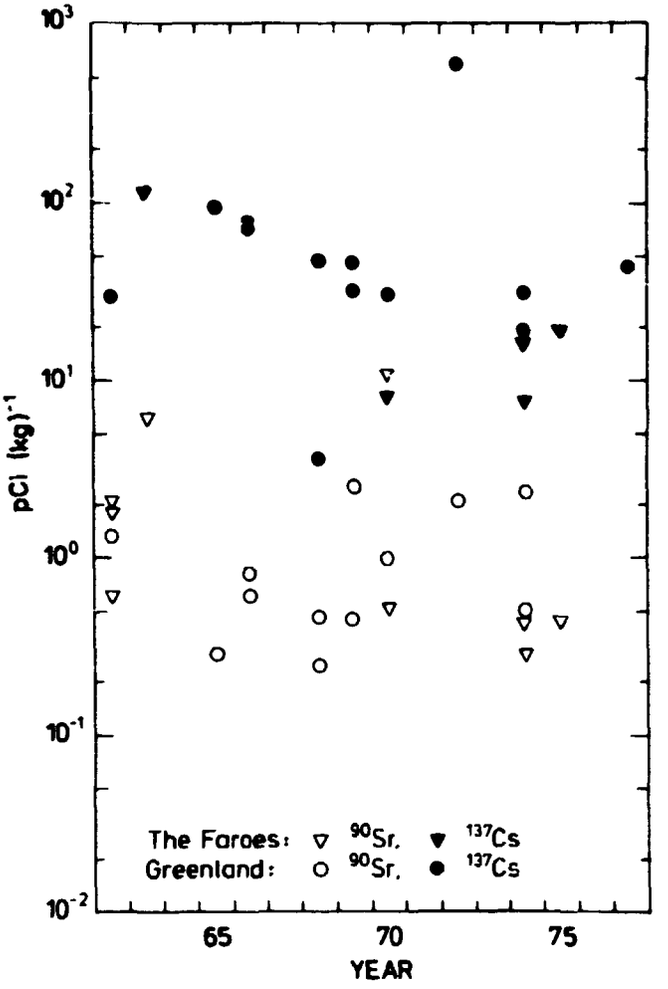

Fig. 3.6.1.1. Strontium-90 and ^{137}Cs in individual samples of whale flesh from animals collected in the Faroes and Greenland 1962-1977 (cf. Table A.1.3.6.1.).

As for other animals, the ^{90}Sr levels in the meat were 1-2 orders of magnitude lower than the ^{137}Cs concentrations, which ranged between approx. 10 and 100 pCi kg^{-1}. Although whales tended to show higher concentrations than seals, the difference between the two animals was not significant. The concentrations showed a decreasing time trend (figs.3.6.1.1. and 3.6.1.2.). The levels were thus approx. 5-10 times higher at the beginning of the sixties than 10 years later. The variabilities among years were relatively high, but as discussed in table B.3.6.1. this was probably the result of an artifact. Two samples of whale meat from 1970 and 1972, respectively, showed exceptionally high levels (1700 and 570 pCi ^{137}Cs kg^{-1}). Had the source of these enhanced levels been Windscale, some ^{134}Cs could have been expected together with the ^{137}Cs (Ku78), but the samples were free of ^{134}Cs; the origin of the high levels was thus unknown.

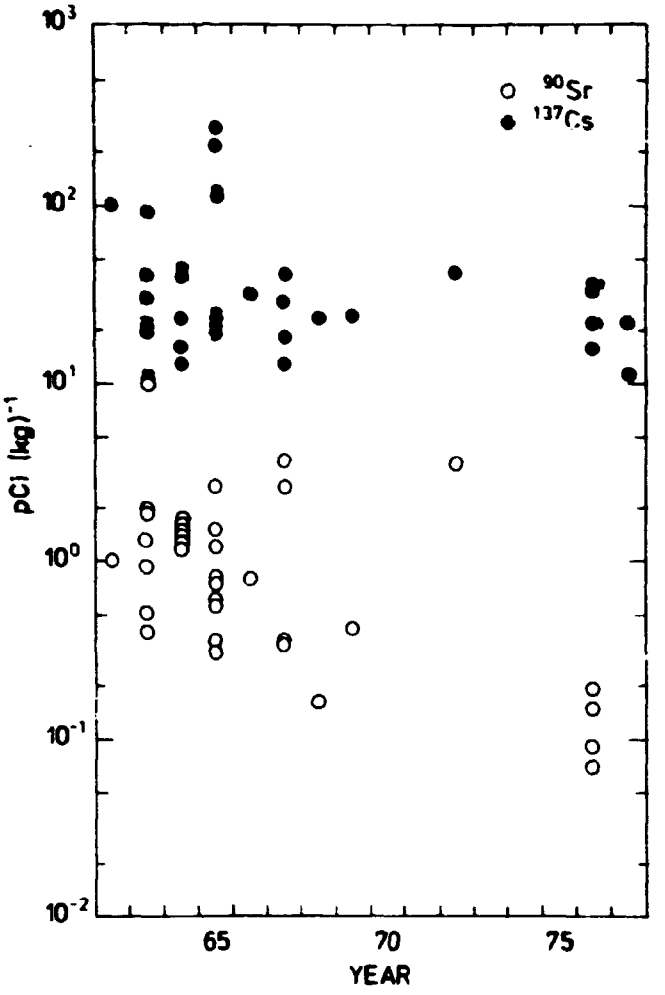

Fig. 3.6.1.2. Strontium-90 and ^{137}Cs in individual samples of seal flesh from Greenland collected in 1962-1977 (cf. Table A.1.3.6.1.).

Prediction models were estimated for Greenlandic seal meat collected in 1963-1969. The radioecological sensitivities became 0.3 pCi ^{90}Sr kg^{-1} y per mCi ^{90}Sr km^{-2} (table C.3.6.1.) and 5 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2} (table C.3.6.2.). In the case of whales, the prediction models were calculated for the Greenlandic as well as for the Faroese environment, because the radionuclide content of these animals does not necessarily originate from the area where they were caught. The radiooecological sensitivities were 5 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2} in the Faroes and 15 in Greenland, and for ^{90}Sr: 0.3 pCi ^{90}Sr kg^{-1} y per mCi ^{90}Sr km^{-2} and 0.8, respectively.

3.6.2. Birds and eggs

In Greenland and the Faroes 20-25% of the "meat and eggs" consumed originate from birds and eggs (RRF62, RRG62), corresponding to approx. 9 kg per capita per year. In Denmark, the annual egg consumption was approx. 11 kg per capita. The birds and eggs consumed in the Faroes and Greenland are derived to a considerable extent from sea-birds. In Denmark, avian food products are mostly derived from poultry.

The ^{90}Sr and ^{137}Cs concentrations in hens' eggs (figs.3.6.2.1 and 3.6.2.2.) from Denmark decreased by an order of magnitude from 1963-1964 to 1974-1975. The variability of the radionuclide concentrations among years (table B.3.6.1.) was approx. 1.3, which was comparable with the variabilities observed for

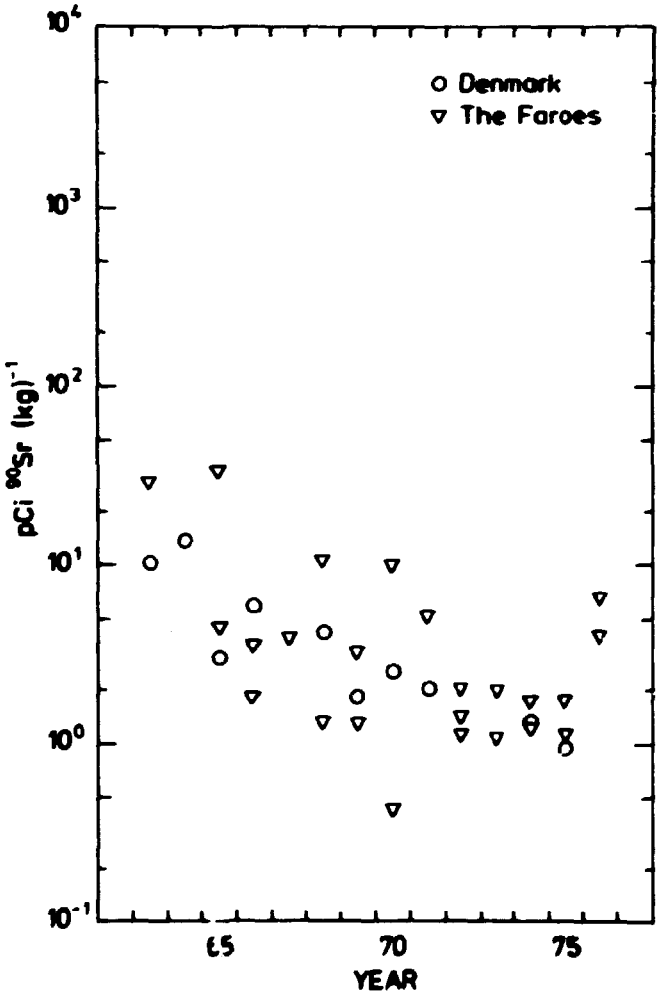

Fig. 3.6.2.1. Strontium-90 in Danish and Faroese hens' eggs (minus the shell) collected in 1962-1976 (Table A.1.3.6.2.).

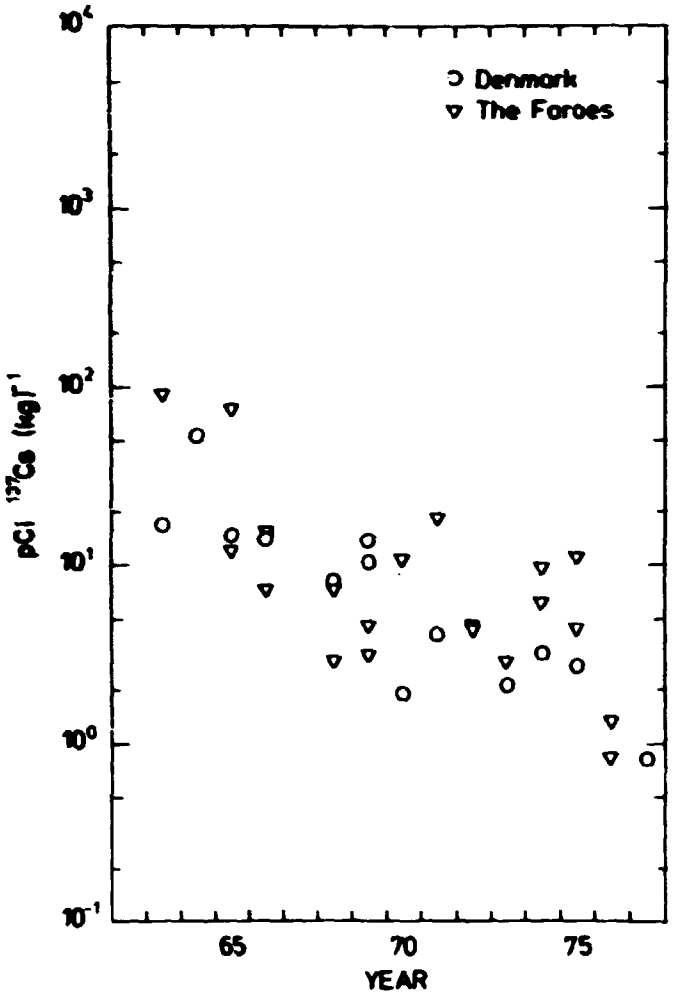

Fig. 3.6.2.2. Cesium-137 in Danish and Faroese hens' eggs (minus the shell) collected in 1963-1977 (Table A.1.3.6.2.).

^{137}Cs in milk (3.2.1.) and meat (3.4.1.). The ^{137}Cs levels in eggs were approx. 3 times the ^{90}Sr levels. The difference between the concentrations of the two nuclides was thus far less pronounced than in the case of meat and fish. Two samples of chicken meat and chicken pluck from 1972 showed a ^{137}Cs concentration 3-4 times higher than that of eggs; this suggested a discrimination against ^{137}Cs in hens' eggs. In the Faroes, the ^{90}Sr and ^{137}Cs concentrations in hens' eggs were similar to those observed in Denmark, because some of the chicken feed (grain) used in the Faroes originates from Denmark.

Samples of sea-birds from the Faroes (Fulmar (Fulmarus glacialis), Guillemot (Uria aalge), Puffin (Fratercula arctica) and Razorbill (Alca torda)) and from Greenland (Brünnich's

Guillemot (Uria lomvia), Black Guillemot (Cepphus grylle) and Eider (Somateria mollissima)) showed ^{137}Cs and ^{90}Sr concentrations similar to those in fish and seals (fig.3.6.2.3.).

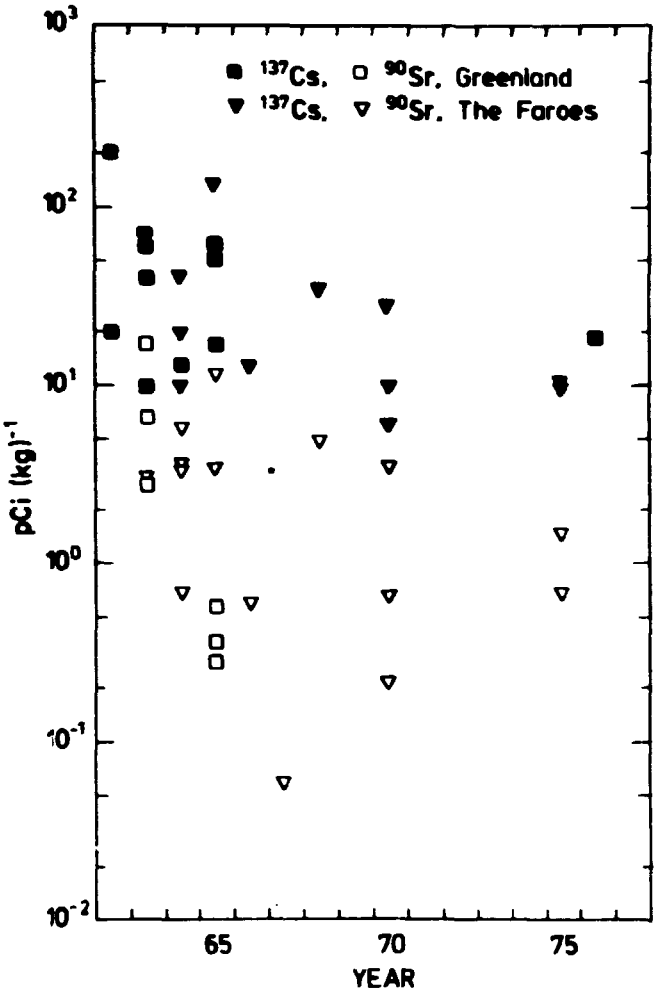

Fig. 3.6.2.3. Strontium-90 and ^{137}Cs in flesh of Faroese and Greenlandic sea-birds (cf. Table A.1.3.6.2.), collected in 1962-1976.

The prediction models for Danish hens' eggs showed transfer factors of 1.3 pCi ^{90}Sr kg^{-1} y per mCi ^{90}Sr km^{-2} (table C.3.6.1.) and 1.8 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2} (table C.3.6.2.). Whereas the radioecological sensitivity to ^{90}Sr was thus close to that observed for beef and pork, the sensitivity to ^{137}Cs was approximately 20 times lower. Hens' eggs are thus an animal product with a low sensitivity to radioactive contamination by ^{137}Cs.

As the material relating to sea-birds was sparse, the data from Greenland and the Faroes were combined by the VAR-3 procedure (cf.B.3.) before the prediction models for the two areas were calculated. The radioecological sensitivity in North Atlantic sea-birds to ^{137}Cs contamination was estimated at 3.5 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2} (table 3.6.2. Nos. 8 and 9), i.e. similar to the sensitivities observed for fish (3.5.2.) and marine mammals.

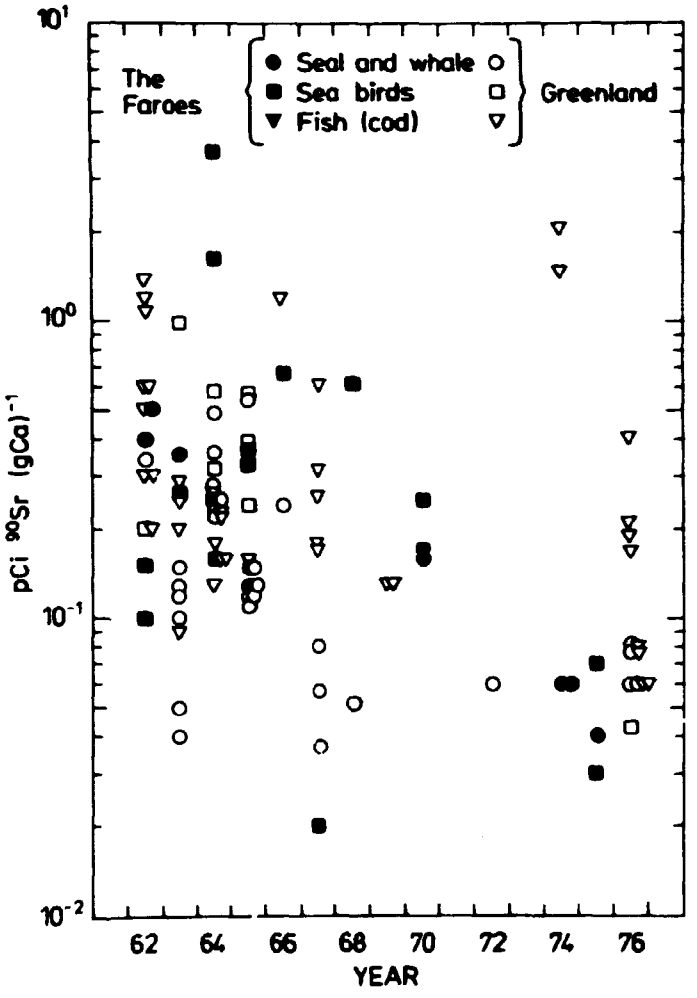

Fig. 3.6.2.4. Strontium-90 in the bones of marine mammals, birds and fish collected in the Faroes and Greenland in 1962-1976.

The ^{90}Sr contents of the bones of marine animals from the Faroes and Greenland (cf. fig.3.6.2.4.) were similar as regards locations and species; but a decreasing trend with time was evident - in the first half of the sixties the levels were about 0.1-1 pCi ^{90}Sr $(g\ Ca)^{-1}$ and by the middle of the seventies they had decreased to 0.05-0.5 pCi ^{90}Sr $(g\ Ca)^{-1}$.

3.7. Conclusions

3.7.1. General

Both the origin and the composition of cattle fodder influence the radioactive contamination of cows' milk. A high proportion of grass results in higher ^{137}Cs levels in Danish milk than those observed in milk from cows on a root diet, but the ^{90}Sr milk levels are less sensitive to the composition of the fodder. In the Faroes the high indirect contamination of the fodder by ^{137}Cs enhances the ^{137}Cs concentrations in the milk. The terrestrial animals used for human consumption are in general herbivores, hence the radioactivity levels in meat are related to the concentrations found in the vegetation making up the animals' fodder. The extreme levels found in lichen are thus reflected in reindeer meat. Salt-water fish and marine mammals contain in general lower radioactivity levels than terrestrial animals; however, as the radionuclide concentrations in the sea are related to the accumulated deposit rather than to the fallout rate, the infinite time-integrated levels in marine animals are not entirely negligible. Apart from milk, animal products are in general low in ^{90}Sr, whereas fish and meat as well as milk are important ^{137}Cs donors to the human diet.

3.7.2. Danish milk

The variabilities among years of ^{90}Sr ($CV_{p\ years} = 0.7$) and of ^{137}Cs ($CV_{p\ years} = 1.5$) in Danish milk were within the ranges of those observed for grain and vegetables. Within years, the ^{90}Sr milk levels were nearly constant from month to month, while the ^{137}Cs concentrations were 2-3 times higher in the summer months than in midwinter. This resulted from the change in the composition of fodder during the year, which influenced the $^{137}Cs/^{90}Sr$ ratio of the cattle feed. The local variability was higher for ^{137}Cs ($CV_{p\ locations} = 0.35$) than for ^{90}Sr ($CV_{p\ locations} = 0.25$). Milk from Lolland-Falster showed a lower $^{137}Cs/^{90}Sr$ ratio than milk from West Jutland, because the fodder from Lolland-Falster contained relatively high amounts of refuse from sugar beet factories, and this material was lower in $^{137}Cs/^{90}Sr$ than, e.g., grass.

On the average, Danish milk products cortained 11% higher concentrations of ^{90}Sr and ^{137}Cs than the milk consumed in Denmark. However, in recent years the increasing consumption of milk from Jutland in east Denmark has reduced the difference.

The radioecological sensitivities of Danish milk were 3.3 pCi ^{90}Sr (g Ca)$^{-1}$ y (or 4 pCi ^{90}Sr l^{-1} y) per mCi ^{90}Sr km^{-2} and 3.4 pCi ^{137}Cs (g K)$^{-1}$ y (or 5.5 pCi ^{137}Cs l^{-1} y) per mCi ^{137}Cs km^{-2}. The sensitivities of milk from Jutland were 1.3 and 1.5 times higher than those of milk from the Islands with respect to contamination by ^{90}Sr and ^{137}Cs, respectively.

Of the ^{90}Sr intake with fodder, the Danish cows secreted 0.2% l^{-1} milk and of ^{137}Cs 0.7% l^{-1}. The "observed ratio" of ^{90}Sr (g Ca)$^{-1}$ in milk and fodder was in the range 0.09 - 0.15.

The radioecological sensitivity of Danish milk to ^{89}Sr contamination was 13% of that of ^{90}Sr. The total infinite time-integrated ^{89}Sr level in milk contaminated by fallout from the 1961-1962 test series was 80% of that of ^{90}Sr.

The transfer factor of ^{131}I from deposition to milk was 0.6 pCi ^{131}I l^{-1} y per mCi ^{131}I km^{-2}. The transfer factor from grass to milk was 0.01 pCi ^{131}I l^{-1} per pCi ^{131}I kg^{-1} dry weight (20% dry matter in grass). Danish milk showed a relatively low sensitivity to contamination with ^{131}I because of the high productivity of the pastures, and also because of the use of relatively large amounts of supplementary fodder, which is often stored food that is free of short-lived nuclides such as ^{131}I.

3.7.3. Faroese milk

Contrary to Danish milk, the variability among years of ^{90}Sr and ^{137}Cs in the Faroes was nearly identical for the two nuclides ($CV_{p\ years} = 0.7$), indicating that indirect contamination was of equal importance for the two nuclides. Faroese milk did not show any marked seasonal variations for ^{137}Cs such as was the case for Danish milk. The local variability was more pronounced for ^{137}Cs ($CV_{p\ locations} = 0.3$) than for ^{90}Sr

($CV_{p\ locations}$ = 0.05). Milk from Tværå thus contained 1.8 times more ^{137}Cs than milk from Thorshavn, while the ^{90}Sr levels were nearly the same.

The radioecological sensitivities of Faroese milk were 9 pCi ^{90}Sr $(g\ Ca)^{-1}$ y (or 10 pCi ^{90}Sr l^{-1} y) per mCi ^{90}Sr km^{-2} and 35 pCi ^{137}Cs $(g\ K)^{-1}$ y (or 60 pCi ^{137}Cs l^{-1} y) per mCi ^{137}Cs km^{-2}. These sensitivities were 2.6 and 10 times higher, respectively, than the corresponding sensitivities of Danish milk. Taking the higher deposition in the Faroes into account, Faroese milk contains nearly 6 times more ^{90}Sr and 20 times more ^{137}Cs than Danish milk. The radioecological sensitivities of Faroese grass suggested that imported fodder reduced the levels in Faroese milk compared to those expected for a pure Faroese grass diet.

3.7.4. Meat from terrestrial animals

The variability among years of ^{137}Cs in Danish beef ($CV_{p\ year}$ = 1.2) was similar to that of milk, and this was also the case for ^{90}Sr ($CV_{p\ year}$ = 0.6). The radioecological sensitivity of beef was 27 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2}, or beef was 5 times more sensitive than milk to contamination with ^{137}Cs. In the case of ^{90}Sr in beef, the radioecological sensitivity was 20 times lower than that of ^{137}Cs. Caesium-137 in pork showed a higher variability among years ($CV_{p\ years}$ = 1.6) than beef, but within the years the variability in pork was lower because of a more constant seasonal composition of the fodder. The radioecological sensitivity of Danish pork was 34 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2}, and for ^{90}Sr the sensitivity was 40 times lower.

Faroese and Greenlandic mutton showed similar variabilities of ^{137}Cs among years ($CV_{p\ years}$ = 0.9). The radioecological sensitivity of Faroese and Greenlandic mutton was estimated at 0.3 nCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2}, i.e. an order of magnitude higher than that of Danish meat. As compared with Faroese milk, Faroese mutton showed a 3 times higher sensitivity to ^{137}Cs contamination. With respect to ^{90}Sr, the sensitivity of mutton was 4 pCi ^{90}Sr kg^{-1} y per mCi ^{90}Sr km^{-2}, or 75 times lower than that of ^{137}Cs.

The variabilities among years of ^{137}Cs and ^{90}Sr in reindeer meat were in the range 1.6 to 1.0, which was higher than the variability of lichen (0.3-0.4), suggesting that lichen was not a dominating constituent of reindeer fodder in Greenland. Reindeer meat from the spring and late winter contained 2-3 times more ^{90}Sr and ^{137}Cs than autumn samples. The radioecological sensitivities of Greenland reindeer meat were 1.5 nCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2} and 15 pCi ^{90}Sr kg^{-1} y per mCi ^{90}Sr km^{-2}, and of reindeer bone 100 pCi ^{90}Sr (g Ca)$^{-1}$ y (or 37 nCi ^{90}Sr kg^{-1} y) per mCi ^{90}Sr km^{-2}. Reindeer showed the highest radioecological sensitivity of the animal samples in this study.

The radioecological sensitivity of musk ox meat from East Greenland was estimated at 0.08 nCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2} and 4 pCi ^{90}Sr kg^{-1} y per mCi ^{90}Sr km^{-2}, which was 20 and 4 times lower, respectively, than the corresponding sensitivities for reindeer.

3.7.5. Fish

The variabilities among years of ^{90}Sr and ^{137}Cs in fish were similar, and they were lower in Denmark ($CV_{p\ years} = 0.3$) than in the Faroes and Greenland ($CV_{p\ years} = 0.5$), in agreement with the variabilities observed for sea water from these locations.

The radioecological sensitivity of cod from Danish waters was 20 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2}, the corresponding sensitivities in Faroese and Greenlandic codfish were 2 and 9, respectively. The transfer factors from sea water to fish flesh were 90, 60 and 40 pCi ^{137}Cs kg^{-1} y per pCi ^{137}Cs l^{-1} y for Danish, Faroese and Greenlandic codfish, respectively, suggesting that the transfer of ^{137}Cs was relatively higher in low salinity water than in high. Due to its dependence upon the accumulated ^{137}Cs in the sea, the radioecological sensitivity of Danish cod was as high as 75% of that of Danish beef. The radioecological sensitivities of fish to ^{90}Sr contamination were in general 1-2 orders of magnitude less than those of ^{137}Cs, in agreement with the observations for terrestrial animals.

3.7.6. Various animals

The radioecological sensitivities of marine mammals (seals and whales) from Greenlandic and Faroese waters were in the range of 5-15 pCi $^{137}Cs\ kg^{-1}$ y per mCi $^{137}Cs\ km^{-2}$ and 0.3 - 0.8 pCi $^{90}Sr\ kg^{-1}$ y per mCi $^{90}Sr\ km^{-2}$, which was similar to the sensitivities of fish and sea-birds from these areas.

Hens' eggs from Denmark showed nearly the same variability among years for ^{90}Sr and ^{137}Cs ($CV_{p\ year}$ = 1.3), which corresponded to the variabilities of ^{137}Cs in milk and meat. Faroese eggs contained concentrations of ^{90}Sr and ^{137}Cs similar to the Danish. The radioecological sensitivity of Danish eggs was 1.3 pCi $^{90}Sr\ kg^{-1}$ y per mCi $^{90}Sr\ km^{-2}$, i.e. similar to that of Danish meat, but in the case of ^{137}Cs the sensitivity of eggs was 20 times lower than that of meat, namely 1.8 pCi $^{137}Cs\ kg^{-1}$ y per mCi $^{137}Cs\ km^{-2}$.

4. MAN - TOTAL DIET AND HUMAN TISSUES

4.1. Introduction

Man is by definition the last link in the human food chain. In health physics man is considered the critical organism and dose limits are established with a view to protect individuals, their progeny and mankind as a whole (Ic77). If man in his natural environment is not at risk from a radiological point of view, it may be anticipated that populations of other organisms in the same environment are also protected against radiation hazards.

In the preceding chapters emphasis has been laid on the two radionuclides whose use of the food chain is of foremost importance as a pathway to man, namely ^{90}Sr and ^{137}Cs. Radiostrontium concentrates in calcified tissues, i.e. in human bone and teeth, while radiocaesium accompanies potassium and thus occurs in soft tissue. Wholebody counting makes possible the in vivo assessment of the ^{137}Cs content in humans, because ^{137}Cs - ^{137}Ba are detectable by NaI scintillation counting, being γ-emitters. In the case of ^{90}Sr it is impossible to determine the radionuclide content with reasonable accuracy by means of an in vivo measurement, because both ^{90}Sr - ^{90}Y are β emitter . Only if there is substantial contamination is an assessment possible by means of a bremsstrahlung measurement of ^{90}Y (E max = 2.3 MeV) (Be64a). Radiostrontium in man must thus be determined either by means of autopsy samples of bone tissue or by the analysis of tooth samples. Excreta and urine (Cz63) may be used for an indirect evaluation of the ^{90}Sr content of the human body. Other methods have been proposed, e.g., analysis of human hair (Ho63). Such methods have, however, not been applied to any large extent because of difficulties in interpreting the results.

As the concentrations of ^{90}Sr and ^{137}Cs in human tissue are

closely related to the diet levels, the study of the concentrations of these radionuclides in the total human diet is warranted. Although some items of food are more important than others as radionuclide donors to the diet, the relative contributions from the various diet components show considerable variations in time (X). In studies of the Danish total diet it was thus considered appropriate to include all ingredients, even those normally considered insignificant as donors of fallout radionuclides.

4.2. Total diet

Men det Allerkonstigste var dog, at naar man holdt Fingeren ind i Dampen fra Gryden, saa kunde man strax lugte, hvad Mad der blev lavet i hver Skorsteen, der var i Byen.

SVINEDRENGEN

But the cunningest arrangement of all was that, if you held your finger in the steam from the pot, you could at once smell what was being cooked on every fire in the town.

THE SWINEHERD

Comprehensive long-term studies of ^{90}Sr - and later of ^{137}Cs - in the total human diet have been carried out since the end of the fifties in the UK (Br58, Ag59), the USA (Ku58, Has60), West Germany (De70), Japan (Na63) and in Denmark (RRD60). The UK studies were discontinued in 1965 (Ag66) and replaced by milk analysis because the ^{90}Sr milk concentrations in the UK seemed to be closely related to the total diet levels.

Several methods have been used to estimate the radionuclide content of the total diet. From the analysis of individual diet constituents collected at the location of production, combined with information on the composition of the human diet, the total intake of the various radionuclides with the diet may be calculated. This method has been applied in studies of the Danish, Faroese and Greenlandic diet. The diet components may, however, also be collected at the location of consumption and pooled into one sample before analysis. The two methods are not necessarily identical. In Danish studies (cf.A.1.), milk has thus been represented by dried milk from seven factories and by fresh milk from 48 towns. In the case of grain products, the radionuclide levels found, using the first method, were calculated from grain samples collected at 10 experimental farms, while in the last method bread purchased in 48 towns was

used. Also the samples of potatoes and some other vegetables, of fruit, meat, fish and eggs originated from different sampling locations and sampling periods in the two methods. Finally, direct sampling of meals from private households or institutions may be used. This method was occasionally applied in the present studies (RRD60, RRD62, RRD64, RRD65).

The composition of the diet may be estimated from statistical information on the consumption of foods (Da57-77), or from interviews concerning food habits in selected population groups (Da68). The composition of the Danish total diet was estimated from a combination of these methods by HOFF JØRGENSEN (RRD62) in 1962. The food habits of a population change with time, however, especially during periods with rapid changes in the socio-economic structure of society. For clarity, the composition of the original diet has been kept unchanged in this study. In table D.4.2.1., however, the 1962 diet is compared with a 1975 diet estimate, and the 1976 concentrations of ^{90}Sr and ^{137}Cs in the various diet components were used as an example of the differences between the compositions of the two diets. The composition of the 1962 diet seemed to underestimate the ^{137}Cs levels in 1976, because the consumption of meat and fish has increased since 1962. Nevertheless, as the potassium intake has also increased, the $^{137}Cs/K$ quotient was nearly unchanged in the two diets. Bread consumption has decreased since 1962 while the consumption of milk has increased. This resulted in a net decrease of Ca and stable Sr in the total diet. The Sr/Ca quotient had consequently decreased only a little from the 1962 to the 1975 diet. The variations in the composition of the diet thus played only a minor role with respect to the $^{90}Sr/Ca$ and $^{137}Cs/K$ ratios, but the total intake of ^{137}Cs may have been underestimated by 10-20% in recent years. Prediction models for future levels should therefore be based on the $^{90}Sr/Ca$ and $^{137}Cs/K$ ratios rather than on the total intakes of the two radionuclides.

4.2.1. Variation with time and location

The variability of ^{137}Cs with time in the Danish total diet was twice as high as that of ^{90}Sr (cf. tables B.4.2.1 and B.4.2.2). As appears from figs. 4.2.1.1. and 4.2.1.2, the maximum con-

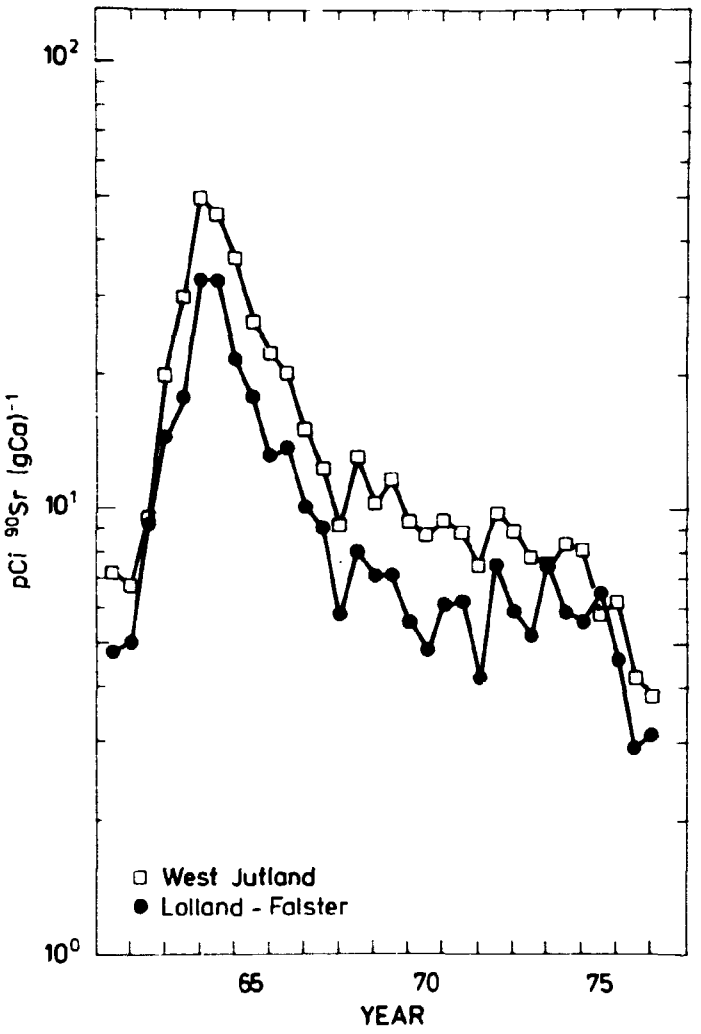

Fig. 4.2.1.1. Strontium-90 in the Danish total diet ("Diet C" cf. Table A.1.4.2.) collected in West Jutland and in Lolland-Falster in 1962-1976; the two areas of the country represented the maximum and the minimum mean values, respectively, for the period of observation.

centrations in the Danish total diet occurred in 1963-1964. Since then the ^{90}Sr levels have decreased by a factor of 10-15, while the ^{137}Cs levels have decreased 25-30 times. The higher time variability of ^{137}Cs (1.29) than of ^{90}Sr (0.60) in the period 1962-1974 was due to the dominating dependence of ^{137}Cs on direct fallout for most of the important ^{137}Cs donors to the diet. The time variability of ^{137}Cs and ^{90}Sr in milk was 1.5 and 0.7, respectively, (1962-1976), and in rye bread the corresponding figures were 1.56 and 0.72 (1962-1974). In fish, the time variability of ^{137}Cs was 0.26 (1963-1973). It was thus evident that fish contributed to a reduction of the time variability of ^{137}Cs in the diet. In the case of ^{90}Sr, the low

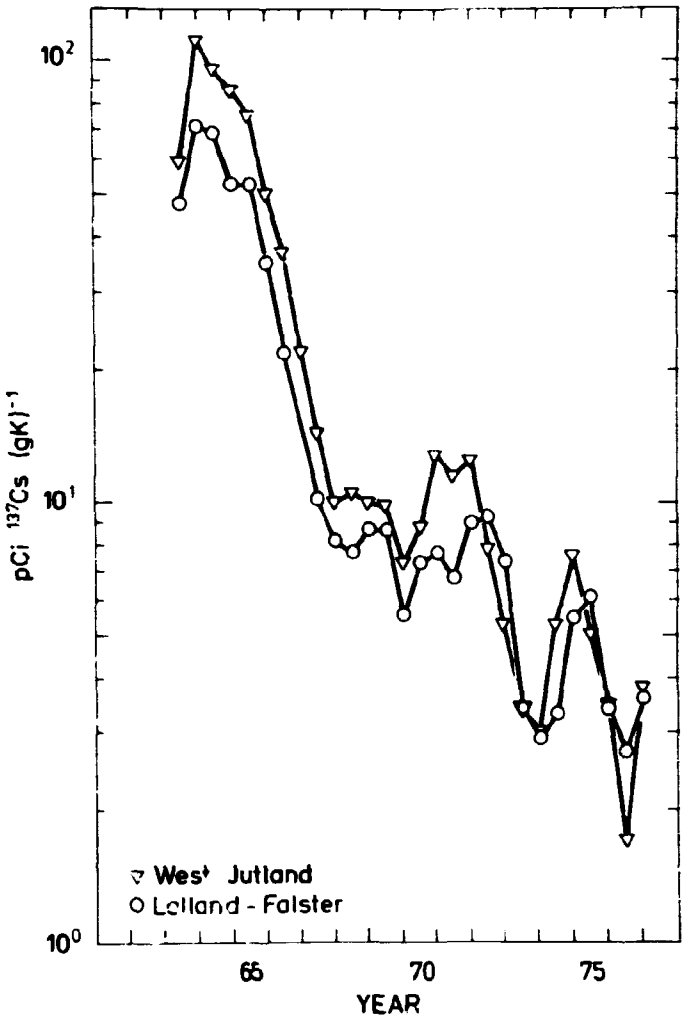

Fig. 4.2.1.2. Cesium-137 in the Danish total diet ("Diet C", cf. Table A.1.4.2.) collected in West Jutland and in Lolland-Falster in 1963-1976.

variability among years in vegetables (table B.2.5.3.) lowered the variability of the total diet.

The local variation followed the general pattern observed for the various components of the Danish diet; the ^{90}Sr and ^{137}Cs levels in Jutland were higher (1.3 times) than the concentrations in the diet from the Islands. In the case of ^{90}Sr, the concentrations in West Jutland were the highest, while those in Lolland-Falster were the lowest (fig.4.2.1.1). In the case of ^{137}Cs, the local pattern changed with time (fig. 4.2.1.2), i.e. the interaction between location and years was significant (table B.4.2.2.). The local variability was 0.16 for ^{137}Cs and 0.13 for ^{90}Sr during 1962-1966. In 1973-1976 it had decreased to 0.12 and 0.10, respectively (table B.4.2.3),

because the transfer of food (e.g. milk) between the various areas of the country had increased. Milk products were mainly responsible for the local variability of ^{137}Cs in the total diet, but meat was also of importance (cf. tables B.3.2.1. and B.3.4.1). The local variabilities of ^{90}Sr and ^{137}Cs in the total diet were lower than the corresponding variabilities for fresh milk ($CV_{p\ (locations\ Sr)} = 0.22$, $CV_{p\ (locations\ Cs)} = 0.27$), and higher than of white bread ($CV_{p\ (locations\ Sr)} = 0.04$, $CV_{p\ (locations\ Cs)} = 0.06$), but similar to those of rye bread (cf. table B.2.3.1). The local variabilities of both ^{90}Sr and ^{137}Cs were also close to that observed for fallout (mCi ^{90}Sr km^{-2}), which displayed a local variability of 0.16 for the state experimental farms during 1962-1974. This was, however, fortuitous as the fallout levels were not the only source of local variation.

4.2.2. Relations and prediction models

As milk is an important constituent of the diet, and plays a special role as a donor of fallout radionuclides such as ^{90}Sr and ^{137}Cs, it has been common practice (Un58-77) to relate diet levels to the milk concentrations of these nuclides. Anovas of the pCi ^{90}Sr $(g\ Ca)^{-1}$ diet/pCi ^{90}Sr $(g\ Ca)^{-1}$ milk ratios (table B.4.2.5) showed a highly significant variation among years (1962-1976) and among the 8 zones. Similar results were obtained by an anova of the corresponding pCi ^{137}Cs $(g\ K)^{-1}$ ratios (table B.4.2.6). The mean ^{90}Sr ratio between diet and milk was 1.44 during 1962-1976, and for ^{137}Cs the corresponding ratio was 1.57. The anovas furthermore showed highly significant interactions between years and locations. It was thus evident that the approach used in the UK of estimating dietary ^{90}Sr levels from milk levels (Ag66) would be inappropriate under Danish circumstances, mainly because milk is less important as a ^{90}Sr donor in the Danish diet than it is in the UK.

As bread and milk contributed 70-90% (X) of the ^{90}Sr in the Danish total diet, it was obvious to consider the diet/milk + bread ratio; however, also this ratio varied significantly with time (1962-1972) as well as with location. There was thus no simple substitute for a sampling of the total diet.

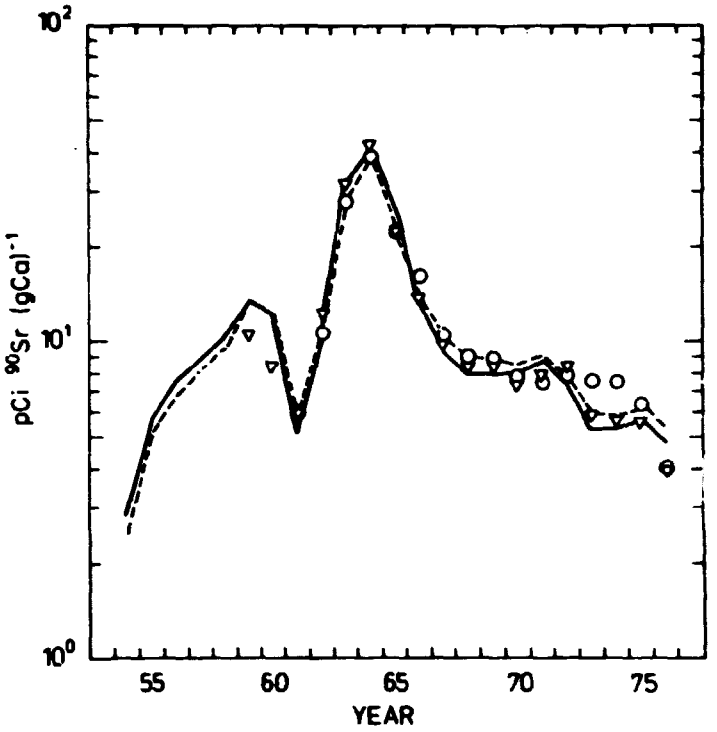

Fig. 4.2.2. Predicted and observed ^{90}Sr levels in the Danish total diet. The dotted curve represents the predicted values for "Diet C" (cf. Table A.1.4.2.) according to Table C.4.2.1. No. 1, and the circles are the corresponding observed values (r = 0.9924***). The unbroken curve represents the predicted values for "Diet P" (cf. Table C.4.2.1. No. 7), and the triangles the corresponding observed values (r = 0.9883***).

Prediction models were calculated for total diet samples (D.4.2.3) ("consumption data") (tables C.4.2.1 and C.4.2.2) and from the data obtained based on single component measurements (RRD59-76) ("production data"). In the following, the two types of sample are called "Diet C" and "Diet P". In fig. 4.2.2. the two diet types are compared with each other and with their respective prediction models. The agreement was generally satisfactory. The radioecological sensitivity for ^{90}Sr in the total Danish diet (unit: pCi ^{90}Sr (g Ca)$^{-1}$ · y per mCi ^{90}Sr km^{-2}), according to the models used in fig.4.2.2., was 6.0 for "Diet C" and 5.0 for "Diet P" (tables C.4.2.1 Nos. 1 and 7). If equations Nos. 2 and 8 in table C.4.2.1. were used, the sensitivities became 4.3 and 5.0, respectively. The mean of these estimates was 5.1 $\pm$ 0.33 (1 SE); the radioecological sensitivity for the diet in Jutland was not significantly different from that in Zealand; both were close to the country mean. This was not surprising, although Jutland milk showed

approx. 1.3 times higher sensitivity than Islands milk (C.3.2.1), because bread, which was the most important ^{90}Sr donor, showed only minor local variations (2.3.1.).

The radioecological sensitivity of ^{137}Cs in the Danish total diet (unit: pCi ^{137}Cs (g K)$^{-1}$ · y per mCi ^{137}Cs km^{-2}) was 4.2 (table C.4.2.2. Nos. 1 and 8) for both "Diet C" and "Diet P". Equation 6 in table C.4.2.2. gave a sensitivity of 4.7 for "Diet P". The mean of the three estimates was 4.4 $\pm$ 0.15 (1 SE); in the case of ^{137}Cs the radioecological sensitivity was 1.08 times higher in Jutland than in the Islands. This difference from ^{90}Sr was not unexpected as milk (and thus presumably also beef) showed a higher sensitivity in Jutland than in the Islands, and also because bread was less important as a ^{137}Cs donor than milk and meat (X).

A comparison between the radioecological sensitivities of ^{90}Sr and ^{137}Cs required a normalisation of the sensitivities to, e.g., pCi (caput)$^{-1}$. According to table D.4.2.1., the annual per caput intakes were 623 g Ca and 1281 g K (1962 diet composition); hence the sensitivities became 3.2 nCi ^{90}Sr per caput per mCi ^{90}Sr km^{-2} and 5.6 nCi ^{137}Cs per caput per mCi ^{137}Cs km^{-2}. The total diet was thus 1.75 times more sensitive to contamination with ^{137}Cs than with ^{90}Sr. As the total ^{90}Sr deposited in Denmark was 73 mCi km^{-2}, and the total ^{137}Cs was 117 mCi km^{-2}, the infinite exposure integrals to the Danish population became 234 nCi ^{90}Sr per caput and 655 nCi ^{137}Cs per caput, i.e. the intake with diet of ^{137}Cs from nuclear weapons fallout was 2.8 times that of ^{90}Sr.

Table 4.2.2. lists the contributions from the various diet components to the radioecological sensitivity of total diet. It appears that the sums of the individual contributions are close to those estimated for the total diet. It should be noted that coffee and tea were not included in table 4.2.2., which means that the sums should probably have been a few per cent higher (2.5.4.). UNSCEAR (Un77) has also estimated the contributions from the various diet components to the total intake of ^{90}Sr and ^{137}Cs in Denmark, as shown in table D.4.2.2. The UNSCEAR estimate was in general agreement with the present one.

Table 4.2.2. Contributions from the individual diet components to the radioecological sensitivity of the Danish total diet.

Component	nCi ^{90}Sr (Cap)$^{-1}$ per mCi ^{90}Sr km^{-2}		nCi ^{137}Cs (Cap)$^{-1}$ per mCi ^{137}Cs km^{-2}		References to the prediction equations
Milk and cheese[1]	0.89		0.94		Tables C.3.2.1 and C.3.2.2
Rye bread	0.91		1.50		Table C.2.3.1 Nos. 1 and 4
White bread	0.24	1.25	0.59	2.18	Table C.2.3.1 Nos. 2, 3 and 5
Grits (oats)	0.10		0.09		Table C.2.3.1 Nos. 10 and 11
Potatoes	0.19		0.28		Table C.2.5.1 Nos. 8, 9 and 10 Table C.2.5.3 Nos. 5, 6 and 7
Leaf vegetables[2]	0.15		0.03		Table C.2.5.1 Nos. 1, 2, 3 and 4 Table C.2.5.3 Nos. 1 and 2 Table C.2.5.5 Nos. 1 and 6
Root vegetables[3]	0.12	0.77	0.01	0.54	Table C.2.5.1 Nos. 5, 6 and 7 Table C.2.5.3 Nos. 3 and 4 Table C.2.5.5 Nos. 4 and 9
Peas and beans[4]	0.05		0.04		Table C.2.5.5 Nos. 2, 3, 7 and 8
Fruit[5]	0.26		0.18		Table C.2.5.1 No. 13 Table C.2.5.3 Nos. 10 and 11 Table C.2.5.5 Nos. 5 and 10
Pork	0.03		1.26		Table C.3.4.1 Nos. 3 and 4 Table C.3.4.2 Nos. 3 and 4
Beef	0.02	0.06	0.50	1.78	Table C.3.4.1 Nos. 1 and 2 Table C.3.4.2 Nos. 1 and 2
Eggs	0.01		0.02		Table C.3.6.1 Nos. 5 and 6 Table C.3.6.2 Nos. 6 and 7
Fish[6]	0.01		0.14		Table C.3.5.1 Nos. 1, 2 and 5 Table C.3.5.2 Nos. 1 and 5
Water	0.00		0.00		Table C.1.4.1 Nos. 7 and 8
Σ	2.98		5.58		

The annual per caput amounts were taken from Table D.4.2.1. (1962 diet composition):

1) The transfer factors for milk products were taken from 3.2.2. assuming that 1 kg milk contained 1.2 g Ca and 1.6 g K and 1 kg cheese contained 8.5 g Ca and 1.2 g K.
2) 96% white cabbage, 4% kale.
3) 50% carrots, 50% onions.
4) 95% peas, 5% beans.
5) 60% apples, 40% soft fruit (strawberry), imported fruit was considered as Danish soft fruit.
6) 50% cod and 50% plaice, and 1 kg fish contained 1 g Ca (as residual bone).

4.2.3. Stable strontium in the Danish diet

The total diet contains stable strontium, which originates essentially from four sources: foods of terrestrial origin receive Sr from the soil taken up by the roots of vegetation; marine animals accumulate stable Sr from sea water; drinking water (1.4.4.) contains Sr as a result of the contact of water with soil and mineral deposits (ground water), but it may also

receive some stable Sr from the influx of sea water; and finally the creta praeparata added to Danish flour contains stable Sr of mineral origin.

Stable Sr in the diet was studied in order to estimate the relative contributions from indirect contamination (root uptake) of ^{90}Sr in the various diet groups. In this case, only the first source of stable Sr mentioned above need be considered. Hence, instead of an annual intake of 1083 mg Sr (cf. table D.4.2.1.), the intake became 243 mg Sr y^{-1} (coffee, tea, beer and wine were not included as drinks). The relative contributions to this total stable Sr intake were 34% from milk products, 27% from grain products, 34% from vegetables and fruit, and 5% from meat and eggs. From the prediction models referred to in table 4.2.2., the contributions to the radioecological sensitivities from the ^{90}Sr accumulated in the soil, i.e. from indirect contamination, may be estimated. According to this calculation, milk products contributed 27% of the overall radioecological sensitivity resulting from indirect contamination of the total diet, grain products 32% vegetables and fruit 39% and meat and eggs 2%. These figures were correlated with those stated above for stable Sr ($r = 0.9313^{*}$), and this supported confidence in the prediction models. It may be noticed that in the case of a wholly indirect contamination, i.e. in the absence of ^{90}Sr in the atmosphere, grain products were no longer the most important contributor of ^{90}Sr to the diet; vegetables and fruit then became the main sources.

The studies of stable strontium may also be used to estimate the discrimination against Sr as compared to calcium when passing from one step in the food chain to another, e.g. from soil to total diet. As long as direct contamination occurs, the observed ratios between pCi ^{90}Sr $(g\ Ca)^{-1}$ in diet and soil are not valid as an estimate for this discrimination. According to table D.4.2.1. and the above assumptions, where creta praeparata, fish and drinks were excluded, the annual calcium intake with the total diet was reduced to 334 g. Hence the stable Sr level in a diet that contained only Sr and Ca originating from root uptake became 0.73 mg Sr $(g\ Ca)^{-1}$. The stable Sr-to-Ca ratio

was determined in cultivated soils from the Danish state experimental farms (table D.1.6.2.3.). The mean content in the ploughing layer was 2.88 ± 0.15 mg Sr (g Ca)$^{-1}$ (± 1 SE for 10 samples). ANDERSEN (An67a) found a mean of 2.90 ± 0.14 mg Sr (g Ca)$^{-1}$ in 21 Danish soils, and he found no difference between the ratios obtained by HCl and by ammonium-acetate extractions. Hence the observed ratio of stable Sr/Ca between diet and soil in Denmark was 0.73/2.9 = 0.25; Ca was thus transported four times as easily from the soil to the diet than Sr.

In 1975 (RRD75) the mean concentration of ^{90}Sr in the ploughing layer (0-20 cm) at the state experimental farms was 102 ± 10 pCi ^{90}Sr kg^{-1} (± 1 SE, 10 locations), the mean calcium content (table D.1.6.2.3) was 2.7 ± 0.7 g Ca kg^{-1}, i.e. the mean pCi ^{90}Sr (g Ca)$^{-1}$ ratio was 38 ± 11. From the observed ratio of 0.25 between soil and diet, the total diet level from indirect terrestrial contamination was estimated at 9.5 ± 2.75 pCi ^{90}Sr (g Ca)$^{-1}$. The measured level was 6.4 pCi ^{90}Sr (g Ca)$^{-1}$ and of this 1.4 pCi was estimated to be due to direct contamination (table C.4.2.1., No. 1), hence only 5 pCi ^{90}Sr (g Ca)$^{-1}$ originated from accumulated fallout. If we corrected this ratio to the above Ca intake from root uptake (334 g y^{-1}), we found $\frac{5 \cdot 620}{334}$ = 9.3 pCi ^{90}Sr (g Ca)$^{-1}$, i.e. in accordance with the estimate of 9.5 based on the ^{90}Sr to Ca ratio in the soil. This agreement suggested that the ^{90}Sr from fallout in the ploughing layer followed the stable Sr, i.e. the availability to the plants of the strontium isotopes did not differ significantly.

ANDERSEN (An67a) has shown that discrimination against radio-strontium relative to Ca probably does not occur during root uptake from Danish soils. Hence the discrimination against Sr from soil to total diet takes place in the later steps of the food chain, e.g. from grass to milk (3.2.) and from wheat to white bread. According to British investigations (Ag62), the ratio of stable strontium to calcium in flour was appreciably lower than that in bran and fine offal; on the average the ratio in flour was 0.71 that of grain.

The stable Sr to Ca ratio was determined in total diet samples collected in 1963-1972 in Denmark. Table B.4.2.4. shows the

anova of the three main effects. The local variation was pronounced. The highest levels occurred in Lolland-Falster, Zealand and Copenhagen in accordance with the high Sr/Ca ratios in drinking water from these localities (1.4.4.). The levels in June were approx. 20% higher than those in December, in accordance with higher Sr/Ca ratios in vegetables in June than later on in the season (2.5.1.).

4.2.4. Faroese total diet

In the Faroes the total diet did not consist of locally produced foods only. Cereals, vegetables, fruit and some milk were imported from Denmark and other countries, especially the U.K. Hence it was not possible to obtain a figure for the radioecological sensitivity of the Faroese total diet in the same way as for the Danish diet. However, in the case of global contamination, such as that from nuclear weapons testing, a sensitivity may be calculated assuming that all the foods imported to the Faroes came from Denmark and that a deposition of 1 mCi km^{-2} in the Faroes corresponded to $\frac{1}{2.08}$ mCi km^{-2} in Denmark (1.3.1.), i.e. the Danish sensitivities were divided by 2.08 to obtain the corresponding Faroese.

Table 4.2.4. shows the results of such a calculation. With respect to ^{90}Sr, the Faroese total diet showed a lower sensitivity than the Danish because the important ^{90}Sr donors were derived from abroad. In the case of ^{137}Cs, however, the radioecological sensitivity of Faroese total diet was nearly three times higher than that of the Danish because of the high sensitivity of Faroese milk, potatoes and mutton to radioactive contamination.

As the fallout rate in the Faroes was 2.08 times that in Denmark, the Faroese total diet contained 1.6 times more ^{90}Sr and 5.7 times more ^{137}Cs than the Danish diet. The radioecological sensitivity of ^{90}Sr and ^{137}Cs in the Faroese total diet may also be related to the congeners Ca and K; the infinite time-integrated pCi ^{90}Sr $(g\ Ca)^{-1}$ and pCi ^{137}Cs $(g\ K)^{-1}$ quotients were respectively 2.3 and 6.2 times those of the Danish diet.

Table 6.2.4. The contribution from the individual diet components to the radioecological sensitivity of the Faroese total diet. (Diet composition according to [illegible] (RBF 62)).

Component	kg y^{-1} (cap)$^{-1}$	g Ca[b] y^{-1} (cap)$^{-1}$	mCi ^{90}Sr (cap)$^{-1}$ per mCi ^{90}Sr km^{-2}		g K[b] y^{-1} (cap)$^{-1}$	mCi ^{137}Cs (cap)$^{-1}$ per mCi ^{137}Cs km^{-2}		References to prediction equations[a]
Faroese milk	109.5	131.4	1.13		175	6.43		Table C.3.3.1 Nos. 1 and 2
Danish milk	36.5	43.8	0.37	1.30	58	2.10	9.54	Table C.3.3.2 Nos. 7, 8, 9, 10, 11, 12
Danish cheese	7.3	62.1	0.13		9	0.01		
Danish rye bread	39.4	63.1	0.27		106	0.65		Table C.3.3.1 Nos. 6 and 8
Danish white bread	59.1	59.1	0.18	0.42	83	3.24	0.74	Table C.3.3.1 Nos. 7 and 9
Danish grits	7.3	24.1	0.05		14	0.05		
Faroese potatoes	91	2.5	0.25		268	1.96		Table C.3.5.1 Nos. 11 and 12
Danish leaf vegetables	12.8	6.2	0.05		42	0.01		Table C.3.5.3 Nos. 8 and 9
Danish root vegetables	7.2	2.3	0.04	0.38		0	4.90	
Danish fruit	14	1.4	0.04		25	0.03		
Faroese mutton	18.5	1.9	0.06		48	1.86		Table C.3.6.1 Nos. 5 and 6
Faroese whale	9.3	0.9	0	0.07	28	0.05	1.87	Table C.3.6.2 Nos. 5 and 6
Faroese birds	4.6	5.0	0.01		14	0.02		Table C.3.6.1 No. 3
Danish eggs	4.6				3			Table C.3.6.2 Nos. 2 and 3
Faroese fish	91	9.1	0.01		301	0.15		Table C.3.5.1 Nos. 3 and 4
								Table C.3.5.2 Nos. 2 and 3
Faroese water[c]	548	2.1	0.06		0	0.02		Table C.1.6.1 Nos. 9, 10, 11
Total Faroese diet		415	2.24		1176	15.32		

a) The contributions from the Danish-produced diet components were obtained from the equations referred to in Table 6.2.2., taking into account the fact that the fallout rate of ^{90}Sr and ^{137}Cs in the Faroes was 2.08 times that in Denmark (cf. the text).

b) The Ca and K contents of the Danish products (except bread) were taken from Table B.6.2.1. The Ca and K levels in Faroese products and in bread were the mean levels in fresh samples (undried) measured throughout the years (RBF 62-76).

c) The ^{137}Cs concentration in drinking water was assumed to be 0.25 times the ^{90}Sr level (Ba 58-78).

The differences between the two diets were enhanced due to the lower Ca and K content of the Faroese diet.

In the case of local airborne contamination limited to the Faroese environment, the imported foods would be free of contamination, and the radioecological sensitivity of the Faroese total diet would be reduced to 1.5 nCi cap^{-1} per mCi km^{-2} for ^{90}Sr. Only a minor reduction of ^{137}Cs would occur because all the important ^{137}Cs donors are of Faroese origin; the sensitivity would in this case be 14.4 nCi cap^{-1} per mCi km^{-2}.

4.2.5. Greenlandic total diet

In analogy with the calculation of the radioecological sensitivity of the Faroese diet, this may be estimated for the diet in Greenland (table 4.2.5.) assuming that the fallout rate measured at Godthåb was representative for the population of Greenland as a whole. A deposition of 1 mCi km^{-2} from global long-lived fallout in Godthåb corresponded to $\frac{1}{0.75}$ mCi km^{-2} in Denmark, and the radioecological sensitivities of Danish-produced foods were consequently divided by 0.75 to estimate the equivalent sensitivities in Greenland.

Table 4.2.5. Contributions to the radioecological sensitivity of the Greenlandic total diet

Component / Food group / Basic product	kg							
Milk								
Cheese								
Rye bread[c]								
White bread[c]								
Grits								
Potatoes								
Vegetables								
Fruit								
Gr. mutton								
Gr. reindeer								
Gr. seal								
Gr. whale								
Gr. seabirds								
Eggs								
Gr. fish[e]	127.8	22.8			421			
Gr. water	540							
Total Greenlandic diet	461				1999			

a) The contributions from the Danish-produced diet components were obtained from the equations referred to in Table 4.1.1., taking account of the fact that the fallout in Greenland was 1.75 times that in Denmark (cf. the text).

b) Included 9.9 kg biscuits (rye, 100% extraction).

c) Included 9.9 kg "white rye bread" (rye, 70% extraction).

d) The Ca and K contents in Danish products were taken from Table 3.4.2.1. The Ca and K levels in Greenlandic products were the mean levels in fresh samples (andried) measured throughout the years 1967-74. The Ca content in fish was estimated from the Faroese samples (NRF 61-74). The Ca content and the $^{137}Cs/^{90}Sr$ ratio in drinking water were estimated to be equal to those of Faroese water (Table 4.2.4.).

e) The transfer factor for ^{90}Sr in fish was estimated as the mean of that for Faroese and Danish cod fish, converted into the Greenlandic fallout rate.

In the case of ^{90}Sr, the sensitivity of the Greenlandic total diet was similar to that of the Danish diet; for ^{137}Cs, the sensitivity of the Greenlandic diet was twice as high, due to the consumption of local, terrestrial animals such as sheep and reindeer.

In the case of local contamination in Greenland, the imported foods would reduce the radioecological sensitivity of the Greenlandic total diet to 0.37 nCi cap^{-1} per mCi km^{-2} for ^{90}Sr and to 6.6 for ^{137}Cs. The Greenlanders could thus be at lower risk as regards ^{90}Sr than the Danish population in a similar situation; concerning ^{137}Cs, the risk would be a little higher than for the Danish population (5.6 nCi cap^{-1} per mCi km^{-2}). However, it should be recalled that diet habits vary considerably in Greenland. Reindeer breeders may thus receive an order of magnitude more ^{137}Cs than seal hunters.

The estimated radioecological sensitivities and the measured calcium and potassium levels in the diet suggested that the infinite time-integrated pCi ^{90}Sr $(g\ Ca)^{-1}$ and pCi ^{137}Cs $(g\ K)^{-1}$ ratios of the Greenlandic total diet were, respectively, 1.1 and 1.7 times the corresponding Danish values, or respectively 0.5 and 0.3 times the Faroese levels.

4.3. Human bone

Dandse skal Du! sagde han, dandse paa dine røde Skoe, til Du bliver bleg og kold! til din Hud skrumper sammen som en Beenrads!

DE RØDE SKOE

"Dance you shall", said the angel, "dance in your red shoes until you are cold and pale, until your skin shrivels up like a skeleton's!

THE RED SHOES

Because of the chemical affinity of ^{90}Sr with calcium, Sr accumulates in bone tissue. Human bone was included at an early stage of the fallout studies. KULP and co-workers (Ku57) performed the first ^{90}Sr fallout measurements of human bone collected in 1955 from the USA, Europe (including Denmark), Africa and the Far East. Later, comprehensive long-term studies have been carried out in the USA by the AEC, Health and Safety Laboratory (Be76), in the UK by the British Medical Research Council (Me73), in West Germany by the University of Kiel (De72), and in Japan by the National Institute of Radiological Science (Na75). Bone programmes have also been accomplished in Australia, Canada, Czechoslovakia, Finland, France, Norway, and the USSR (Un77).

In Denmark, the initial ^{90}Sr analyses of human bone were performed at the University of Copenhagen (Le62) in 1958. The

samples were mostly derived from stillborn infants and children in the Copenhagen area. Countrywide sampling of human bone from all age groups began in 1961 (RRD61). The programme was initiated in co-operation with the Danish National Health Service and carried out by Risø.

The bone samples (vertebrae) were obtained from the institutes of forensic medicine at Copenhagen and Aarhus. Most samples were derived from the victims of accidents and could thus be considered to represent a random and unbiased sample of the population. However, for certain age groups (infants and old people), such samples are relatively few and for these groups samples were mostly derived from the victims of diseases. However, there has been no indication that such bone samples differ with respect to ^{90}Sr from the samples obtained from accident victims (Ha58-78).

4.3.1. Variation with time, age and location

The anovas of human vertebrae showed a significant variation with time for all age groups. The maximum occurred for stillborn

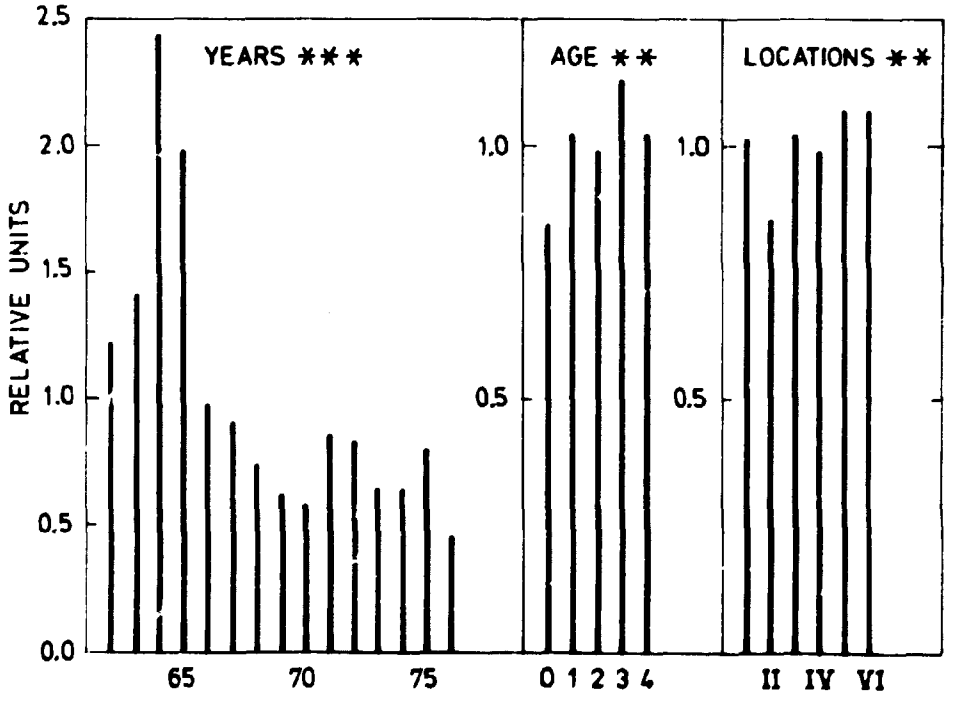

Fig. 4.3.1.1. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in Danish infant bone (0-4 years old) collected in 1962-1976 in zones I-VI (cf. Fig. A.1.4.2.1. and Table A.1.4.3.). The bars show the levels relative to the grand mean 2.8 pCi (g Ca)$^{-1}$ (= 1 on the relative scales).

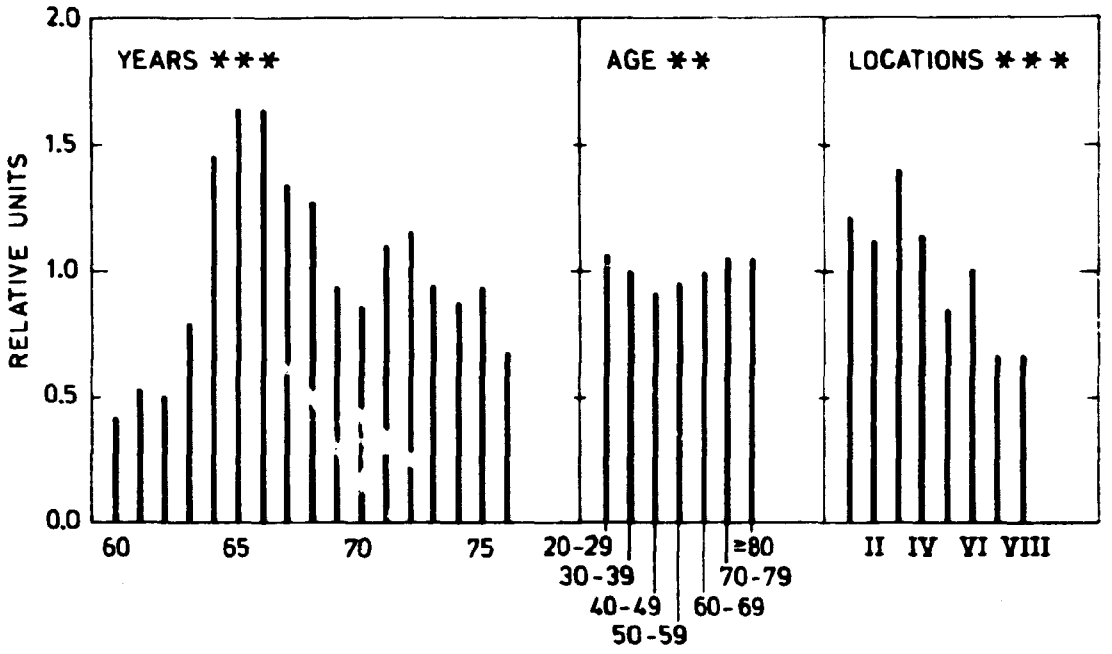

Fig. 4.3.1.2. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in Danish adult bone (20-90 years old) collected in 1960-1976 in the eight zones (cf. Fig. A.1.4.2.1. and Table A.1.4.3.). The bars show the levels relative to the grand mean 1.35 pCi (g Ca)$^{-1}$ (= 1 on the relative scale).

children in 1964, for infants in 1964 (fig.4.3.1.1.), for children in 1965 and for adults in 1965-1966 (fig.4.3.1.2.). As compared with the diet, which peaked in 1963-1964 (fig. 4.2.1.1), the delay was evident and reflected the rate of turnover for Sr in bone, which decreases with age (Be77b). The highest concentrations were found in bone of infants (6-12 months old) and the lowest in bone of adults (> 30 years) and newborn children. In recent years the differences among age groups have become less pronounced due to the decreasing ^{90}Sr concentrations in the diet. This reduces the ^{90}Sr levels in infant bone more rapidly than in the bone of adults and newborn children, the bone levels of the latter also being influenced by the adult ^{90}Sr bone levels (cf.4.3.2.). Finally, adult human bone levels in Denmark varied significantly with location (fig. 4.3.1.2.). Jutland showed approximately 20% higher levels than the Islands, which was compatible with observations of the diet, that contained about 30% more ^{90}Sr in Jutland than in the Islands.

The variability among years (table B.4.3.3.) for the period 1961-1976 was approx. 0.4, and it differed only little between the various age groups. The time variability was a little higher for infants ($CV_{p\ years} = 0.45$) than for children between 5 and 11 years old ($CV_{p\ years} = 0.30$), which reflected the higher turnover rate for ^{90}Sr in infant bone. As expected, the time variability of bone was somewhat lower than that of diet ($CV_{p\ years}$) ~ 0.6). Also as regarded local variability, bone showed lower values ($CV_{p\ locations} \sim 0.1$) than diet ($CV_{p\ locations} = 0.14$). The lesser local variability for bone may partly result from the relocation of people during the period of observation.

Because the bone samples consisted of vertebrae they may not necessarily have been representative of ^{90}Sr in the whole skeleton. Studies in the UK (Br61) have shown that the Sr concentration in children and adolescents was nearly uniform among and within the bones analyzed, thus the ^{90}Sr concentration in the vertebrae of these age groups was essentially equal to that of the whole skeleton. For adults, vertebrae levels may, however, differ from those of the total skeleton because spongy bone such as the vertebrae have a relatively more rapid turnover rate than ivory bones such as, e.g., the femoral diaphysis (Br70). RIVERA (Ri64) found in two human skeletons from 1960 and 1961 that the pCi ^{90}Sr $(g\ Ca)^{-1}$ ratio was 1.8 times higher in the vertebrae than in the entire skeleton. During periods with increasing ^{90}Sr levels in the diet, vertebrae bone will generally show higher concentrations of ^{90}Sr than the skeleton as a whole, but with decreasing levels the situation may reverse.

The average ratio between males and females with respect to ^{90}Sr concentrations in bone tissue was 1.11 in adult bone; but anovas showed no significant difference between the two sexes.

4.3.2. Relations and prediction models

The observed ratio between Sr and Ca in bone and diet ($OR_{bone/diet}$) has been determined by measurements of the stable Sr-to-Ca ratios in the two types of sample. RIVERA (Ri63a) found a mean ratio of 0.16. He analyzed samples of adult bone and of diet

from New York, Chicago and San Francisco, and, although the stable Sr in the three diets differed by a factor of two, he found the same observed ratios for the three locations. The observed ratio between Sr/Ca in Danish vertebrae and diet was 0.12 $\pm$ 0.02 (1 SE, 12 samples), i.e. not significantly different from the findings of RIVERA. Other authors (Br61) though have found $OR_{bone/diet}$ close to 0.25; it has not been clarified whether these differences were due to artefacts or variations in dietary and metabolic factors. It should, however, be noted that the UK study (Br61) was performed on femur shafts, while RIVERA used vertebrae. In another study (Ri63b) RIVERA found the stable Sr concentration of the vertebrae ash to be 0.79 times that of the femur shaft, which may suggest a higher Sr/Ca ratio in the femur than in vertebrae. The $OR_{bone/diet}$ seemed to increase for the youngest age groups; in the first year of life there thus seemed to be very little discrimination by the infant against Sr relative to Ca, i.e. the OR was close to unity (Co66b).

Various prediction models for ^{90}Sr in bone have been proposed (Ri66) throughout the years. One of the more simple and yet adequate models has been that developed by the Health and Safety Laboratory (Be77b):

$$B_n = cD_n + g \sum_{m=o}^{\infty} D_{n-m} e^{-m\lambda} \qquad \text{(Eq.4.3.1)}$$

where

B_n : pCi ^{90}Sr (g Ca)$^{-1}$ in vertebrae in the year n

D_n : pCi ^{90}Sr (g Ca)$^{-1}$ in diet from midyear in the year n-1 to midyear in the year n

c : short-term retention of ^{90}Sr in bone

g : long-term retention of ^{90}Sr in bone

$1-e^{-\lambda}$: effective removal rate for ^{90}Sr in bone including radioactive decay ($\lambda_{Sr-90} = 0.025\ y^{-1}$).

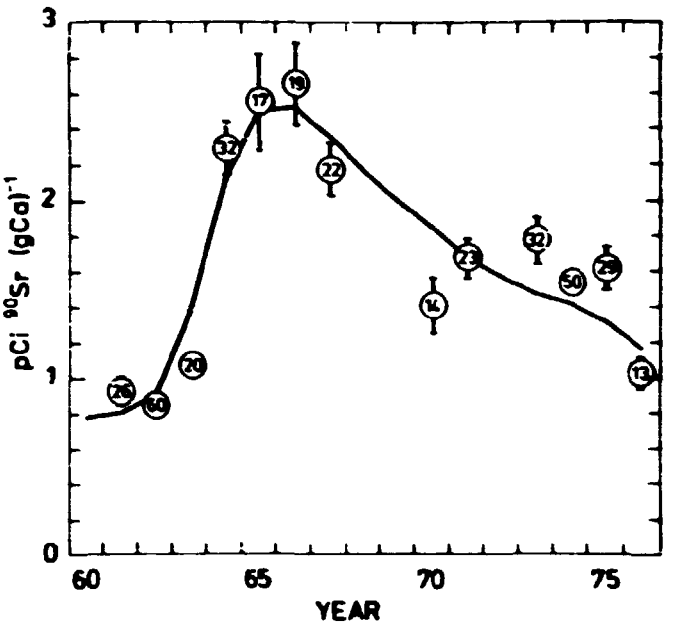

Fig. 4.3.2.1. Strontium-90 in adult human bone from Jutland. The curve represents the predicted values according to:

$$pCi\ ^{90}Sr\ (g\ Ca)^{-1}_{bone(i)} = 0.018 \cdot pCi\ ^{90}Sr\ (g\ Ca)^{-1}_{diet(i)} + 0.028 \sum_{m=1}^{m=\infty} pCi\ ^{90}Sr\ (g\ Ca)^{-1}_{diet\ i-m} \cdot e^{-0.23m},$$

the circles indicate the observed values ± 1 SE, the numbers in the circles the number of samples. All individuals included were at least 30 years old and all were born before 1935.

In figs.4.3.2.1 and 4.3.2.2., Eq.4.3.1 has been applied, with minor modifications, to the adult bone and diet data from Jutland and the Islands, respectively.

Considering a steady state where the fallout rate, and thus the diet and bone levels, has become constant, the observed ratio between bone and diet became:

$$OR_{(bone/diet)\ steady\ state:}\ c + \frac{g}{\lambda - \lambda_{Sr-90}} . \qquad (Eq.4.3.2.)$$

For adult bone, the OR according to (Eq.4.3.2.) should be equal to that observed for mg Sr $(g\ Ca)^{-1}$ in bone and diet. The Danish data (mean of Jutland and Islands data) gave OR = 0.16 (1 SE: 0.005), which was compatible with the above estimate based on stable Sr (Ri63a). However, the US bone data (Be77b) gave an OR of 0.31*), i.e. nearly twice the Danish value. UNSCEAR

*) A recent recalculation has given a figure of 0.10.

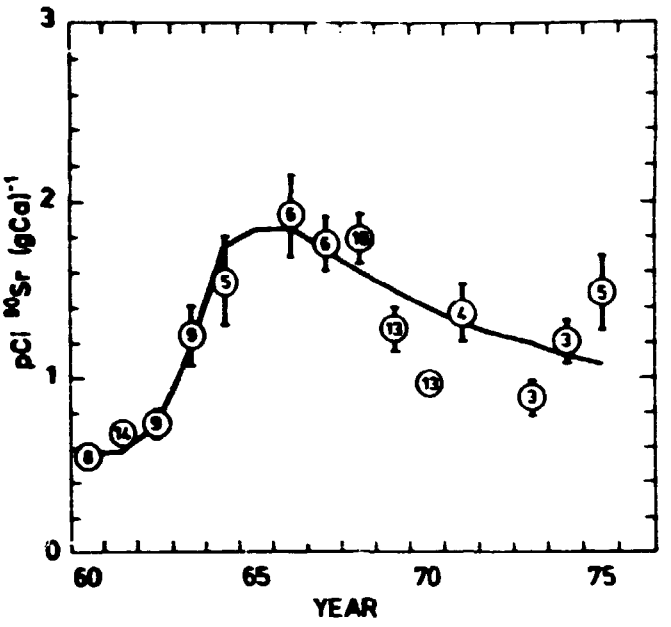

Fig. 4.3.2.2. Strontium-90 in adult human bone from the Islands. The curve represents the predicted values according to:

$$pCi\ ^{90}Sr\ (g\ Ca)^{-1}_{bone(i)} = 0.025\ pCi\ ^{90}Sr\ (g\ Ca)^{-1}_{diet(i)} + 0.022 \sum_{m=1}^{m=\infty} pCi\ ^{90}Sr\ (g\ Ca)^{-1}_{diet\ i-m} \cdot e^{-0.182m};$$

(cf. the caption to Fig. 4.3.2.1).

(Un77) has estimated the transfer factor P_{34} from diet to bone at 0.12, which is reasonably close to an observed ratio of 0.16.

The turnover rate (T) of Sr in adult vertebrae was:

$$T = (1-e^{-\lambda}) - 0.025 \qquad \text{(Eq.4.3.3.)}$$

For bone from Jutland, T = 0.18 y^{-1}, and from the Islands, T = 0.14 y^{-1}, i.e. a mean of 0.16 y^{-1}. In adult vertebrae from New York, BENNETT (Be77b) found a removal rate of 0.23 y^{-1}, while he observed 0.15 y^{-1} for San Francisco adult data. Our data showed that bone from the 20-29 year-old age group displayed higher ^{90}Sr concentrations than the older groups, and a faster turnover rate in the 20-29 year-old group, as observed in D.4.3.2., was to be expected. The Danish adult group consisted of people born before 1935, and who were more than 29 years old, which implied that all members of the sample population had been older than 19 years when ^{90}Sr was introduced

into the human biosphere in 1953-1954 (cf.D.4.3.2.). The US material, included only samples of subjects age 20 years or older in 1954. Thus both the Danish and the US adult materials represented adult metabolism for the entire period of contamination.

Figure 4.3.2.3. shows the $OR_{(new\text{-}born/adult\ diet)}$ based on Danish samples of bone and diet collected since 1963. As the foetus receives a substantial part of its ^{90}Sr (and Ca) from the skeleton of the mother (Bo72) during the last part of pregnancy, the ^{90}Sr concentrations of the foetal skeleton will in periods with decreasing diet levels be relatively higher, as compared with the diet levels, than during periods with increasing diet concentrations of ^{90}Sr. This is because the turnover rate in the maternal skeleton acts as a moderating factor on the foetal ^{90}Sr concentrations. Hence the observed ratios as seen in fig. 4.3.2.3. increased with the decreasing ^{90}Sr diet levels (cf. fig. 4.2.1.1.).

The radioecological sensitivity of human bone to contamination from ^{90}Sr was estimated from the prediction models shown in table C.4.3.1; the models (Nos. 2,4,6,8,10,12 and 14) that assumed a single exponential decay of ^{90}Sr gave average sensitivities which were only 60% of those obtained with the double exponential decay. The transfer factor P_{234} (from fallout to adult bone) was calculated from 4.2.2, where P_{23} (from fallout to diet) was estimated at 5.1 ± 0.33 pCi ^{90}Sr $(g\ Ca)^{-1} \cdot y$ per mCi ^{90}Sr km^{-2}, and from figs.4.3.2.1. and 4.3.2.2, where P_{34} (from diet to bone) was estimated at 0.16 ± 0.005 pCi ^{90}Sr $(g\ Ca)^{-1} \cdot y$, hence $P_{234} = 5.1 \cdot 0.16 = 0.82 \pm 0.06$. According to equations (13) and (14) in table C.4.3.1, P_{234} was 1.05 and 0.59, respectively. In UNSCEAR's latest report P_{234} is estimated at 0.7 for the northern hemisphere (Un77). This estimate was 30% higher than UNSCEAR's previous one from 1972 (Un72). Hence it was a matter of importance which model was used for the prediction of ^{90}Sr in bone. The best available estimate may be the mean of the present three estimates: 0.82, 1.05 and 0.59, i.e. $P_{234} = 0.82 \pm 0.13$ (1 SE), which is thus 20% higher than the latest UNSCEAR estimate.

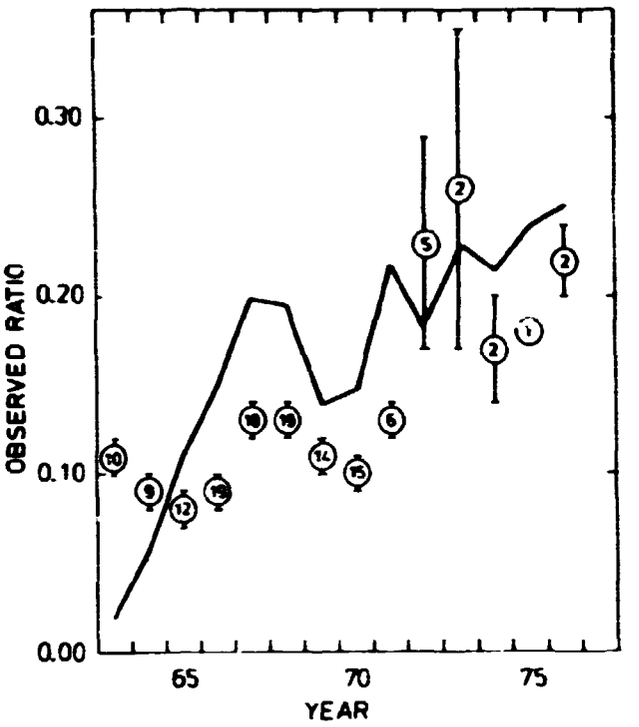

Fig. 4.3.2.3. The observed ratios between pCi ^{90}Sr (g Ca)$^{-1}$ in bone from newborn Danish infants and the corresponding adult diet from the same zone and period as the bone. The number of ratios included in the means from each year is shown in the circles and ±1 SE is indicated. For comparison, the curve shows the observed ratios between pCi ^{90}Sr (g Ca)$^{-1}$ in adult vertebrae and diet.

As regards the radioecological sensitivities of the various age groups, it appeared that the most sensitive group was that covering the first year of life. The sensitivity of this group was approx. 75% higher than that of the adult group. Children were in general more sensitive to ^{90}Sr contamination than adults, except newborn infants whose sensitivity was similar to that of adults.

The observed ratio (OR) between pCi ^{90}Sr (g Ca)$^{-1}$ in bone of Danish newborn infants and Danish diet was 1.04/5.1 = 0.20 according to the radioecological sensitivities of the two samples. A few samples of bone from newborn infants were obtained from the Faroes during 1965-1976 (RRF62-76). The radioecological sensitivity of these samples was estimated at 0.62 pCi ^{90}Sr (g Ca)$^{-1}$ y per mCi ^{90}Sr km^{-2}, (table C.4.3.1 (Nos. 15 and 16) and $OP_{bone/diet}$ was estimated from table 4.2.4 as

0.62/5.4 = 0.12. This was lower than the Danish estimate, but not incompatible with the observations in fig. 4.3.2.3. and with the range 0.1 to 0.2 of $OR_{(newborn\ bone/maternal\ diet)}$ given in the literature (Be77b).

As compared to other western diets, the Danish diet is relatively rich in calcium due to the addition of creta praeparata to the flour. Creta praeparata has been added to Danish flour since 1958 to compensate for the loss of calcium caused by the phytic acid found in cereal products. As demonstrated by COMAR (Co57b), phytate would not influence the discrimination between Sr and Ca, because the solubility of the phytates of the two elements is nearly the same (Han60); and CARR et al. (Ca62) demonstrated that the radiostrontium in wholemeal wheat bread and in milk in the human diet was approximately equally available. In other words, the ratio between Sr and Ca in the organism does not depend on the presence of phytates. Hence, the creta praeparata in the Danish diet has undoubtedly reduced the doses from the relatively high ^{90}Sr intake from rye bread. However, as shown by KORNBERG (Ko59) and other authors (Ha64, Sp67), the reduction in ^{90}Sr uptake is not necessarily proportional to the increase in calcium intake because the observed ratio between bone and diet may vary with the calcium intake.

4.4. Human teeth

Hun kunde huske, jeg fik den første Tand og Familieglæden herover. Den første Tand! Uskylds Tand, skinnende som en lille hvid Mælkedraabe, Mælketanden.

TANTE TANDPINE

She could remember me cutting my first tooth and how delighted the family was The first tooth! The tooth of innocence, shining like a little white drop of milk: the milk tooth.

AUNTIE TOOTHACHE

The ^{90}Sr concentrations in teeth are of no direct interest as a source of internal contamination of the human body. In the present study shed deciduous teeth have been used as a substitute for infant bone, because such bone samples have generally been unavailable from the Faroes and Greenland. To interpret the results of the tooth measurements, the ^{90}Sr concentrations in corresponding samples of Danish shed deciduous teeth and infant bone have been used. The preliminary results of the tooth studies (XI, XII) indicated that ^{90}Sr in shed

deciduous teeth may be considered as a measure of the ^{90}Sr bone level of the 1-year-old tooth donor. This implies, among other things, that the shed deciduous tooth has not exchanged significant amounts of ^{90}Sr with the environment since the calcification of the tooth, i.e. that the turnover rate of ^{90}Sr in the shed deciduous tooth crown after formation is insignificant (D.4.4.1.).

4.4.1. Variation with time, type of tooth and location

The ^{90}Sr concentration in deciduous teeth has shown a pronounced variation with time. At the beginning of the fifties when the fallout rate was negligible, the tooth levels were nearly zero (XI). The maximum occurred in teeth from children born in 1963, i.e. in the cohort that showed the highest bone levels as

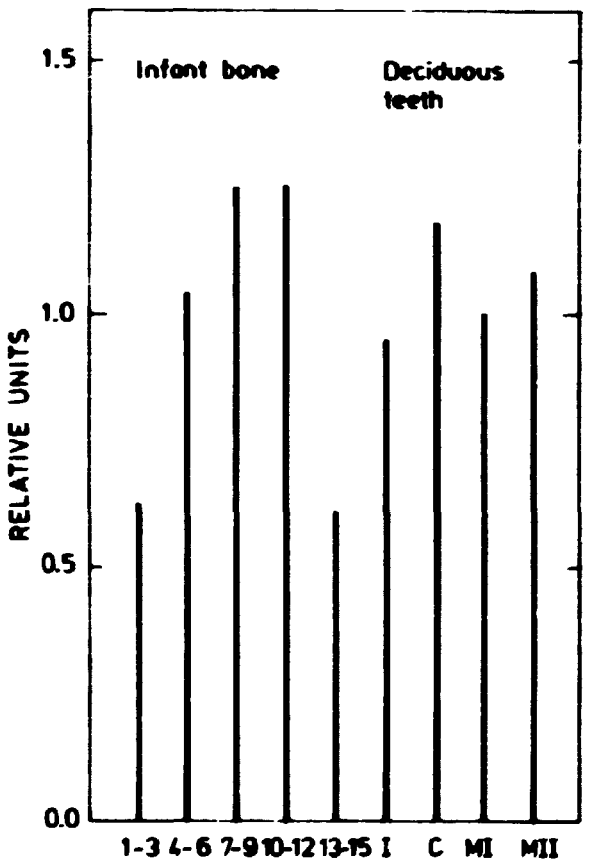

Fig. 4.4.1. The relative pCi ^{90}Sr $(g\ Ca)^{-1}$ concentrations in infant bone (1-3 months, 4-6 months, etc.) and deciduous teeth: incisors, cuspids, first and second molars, collected in 1962-1970 in Denmark (cf. text). The grand mean was 4.5 pCi $(g\ Ca)^{-1}$ (= 1 in the relative scale).

1-year-old infants in 1964. Since then, the levels have been decreasing and deciduous teeth from children born in 1967-1969 contained approx. one third of the maximum level.

Incisors have, in general, contained less and second molars a little more than the mean level of all types of tooth, but in the Faroese and Greenlandic materials the differences among types of tooth were not significant. Teeth from Jutland contained approx. 20% higher ^{90}Sr levels than teeth from the Islands, which finding was in agreement with observations for human bone (4.3.1.).The Greenlandic material contained nearly the same levels as those found in the Danish, but the Faroese tooth levels were significantly higher, as was expected from the elevated ^{90}Sr concentrations of Faroese milk (3.3.1.). The variability among years was the same for teeth from Denmark, the Faroes and Greenland (table B.4.4.2.).

4.4.2. Relations and prediction models

As the ^{90}Sr tooth studies aimed to obtain an estimate of the ^{90}Sr concentrations in infant bone where and when bone samples were scarce or unavailable, mutual ^{90}Sr measurements were carried out on Danish infant bone and corresponding shed deciduous teeth from 1962 to 1970. A comparison could thus be made for this period. An anova showed significant variation among sample types (bone of infants 1-3 months and 12-15 months old contained less ^{90}Sr than the other samples), as well as among years, but no interaction between samples and years. Figure 4.4.1. shows that the ^{90}Sr concentration in shed deciduous teeth of a tooth donor born in the year (i-1) corresponded to that found in his bone when he was around 1 year old in year (i). If the prediction model for 4-12 month-old infant bone (table C.4.3.1., No 3) was used to predict the tooth levels in children born in 1952-1969, and if these predicted values were compared with those actually observed, the correlation coefficient: $r = 0.884$ was highly significant and the mean ratio between the predicted and the observed values was 1.14 ± 0.08 (1 SE), i.e., not significantly different from unity. In a similar way one may compare the measured 4-12 month-old infant bone levels from 1962-1976 with those predicted from

the tooth prediction model (table C.4.4.1, No 1). In this case $r = 0.889$ was also highly significant and the mean ratio was 0.95 ± 0.07. The ^{90}Sr concentrations in shed deciduous teeth may thus be used as an estimate of the bone level of the one-year-old (4-12 month) tooth donor. This has earlier been suggested (XII), but at that time there were too few parallel tooth and bone samples to confirm the hypothesis.

In table C.4.4.1. the radioecological sensitivities of shed deciduous teeth have been calculated. It appears that the sensitivity for Danish teeth was 0.89 pCi ^{90}Sr $(g\ Ca)^{-1}$ y per mCi ^{90}Sr km^{-2}, which was lower than the sensitivity estimated for infant bone (4-12 month-old: 1.1 - 1.7). The reason may be that the tooth material did not represent the later years (1971-1976), when the importance of indirect contamination from the accumulated fallout increased relative to that from direct contamination. The relatively higher uptake of ^{90}Sr from the soil improved the estimate of the soil factors in the prediction models. If the ^{90}Sr results for tooth and infant bone (4-12 month-old) were pooled for the period 1951-1976, and new prediction models (table C.4.4.1., Nos. 5 and 6) were calculated, the radioecological sensitivities became identical to those in 4-12 month-old infant bone (fig.4.4.2.).

The radioecological sensitivity of Faroese deciduous teeth was approx. 1.2 times that of Danish teeth (table C.4.4.1., Nos. 3 and 4). This indicated that Faroese milk played no dominating role as a ^{90}Sr donor to Faroese deciduous teeth as the radioecological sensitivity of ^{90}Sr in Faroese milk was 2.6 times higher than that of Danish milk (3.2.2. and 3.3.2.). Faroese children apparently consumed foods relatively low in ^{90}Sr as compared to Faroese milk; Danish imported milk, human milk, Danish bread, fruit and vegetables would all lower the Faroese infant bone levels relative to a pure Faroese milk diet (cf. 4.2.4.). As fallout in the Faroes was 2.08 times that in Denmark, the Faroese teeth (and infant bone) contained $1.2 \cdot 2.08 = 2.5$ times the Danish levels, and the doses from ^{90}Sr to Faroese infant bone have therefore been 2.5 times the Danish doses. This estimate was lower than the previous one (XI, XII),

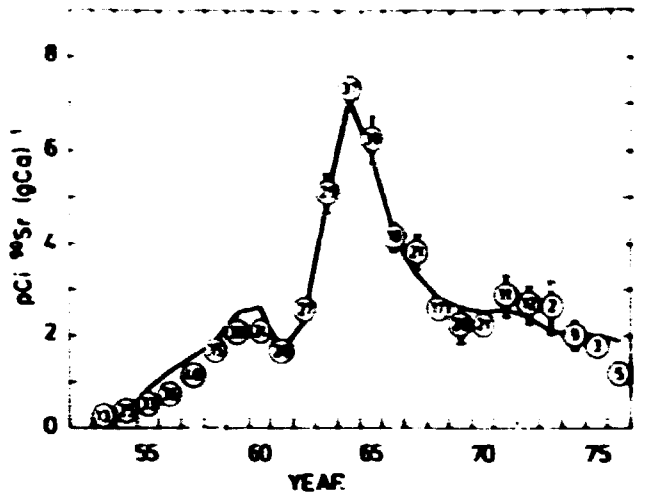

Fig. 4.4.2. Predicted and observed ^{90}Sr levels in Danish infant bone and deciduous teeth. The abscissa is the year of death of the bone donor and the year when the tooth donor completed his first year. The number of samples included is shown in the circles and ±1 SE is indicated. For 1959-1961 the samples consisted of teeth only, during 1962-1970 both teeth and bone were included, and since 1971 bone only. The curve is the predicted values according to Table C.4.4.1. No. 5 ($r = 0.9825^{***}$).

which was estimated as four times the Danish levels. However, similar to the Danish "pure tooth" model, the Faroese model may have underestimated the radioecological sensitivity due to incomplete data from later years.

The tooth samples were the only environmental samples in the present material that covered the period of the first global fallout, i.e. the beginning of the fifties. As the measurements of fallout and of accumulated ^{90}Sr in the soil were very sparse and incomplete from these years, it was difficult to obtain reliable prediction models based on data from this period. To test the influence of the estimated fallout data for the years prior to 1961, the prediction model for teeth was calculated only from the data collected after 1961. This model, however, came very close to the model using all data, thus the potentially unreliable fallout data from the beginning of the fifties did not seem to have much influence on the tooth models. From fig.4.4.2. it appears that the predicted tooth levels in the fifties were generally higher than the observed levels.

This suggests a minor overestimate of the ^{90}Sr fallout levels in this period. Earlier estimates of the fallout in the fifties were lower than the present one; however, there were other reasons for preferring the present estimate (cf.D.1.3.2.) and the observations for teeth do not justify any adjustments.

4.5. The human body

Hun tog den lille Gerda om Livet og sagde: "De skal ikke slagte Dig, saalænge jeg ikke bliver vred på Dig!
SNEDRONNINGEN

She put her arm round little Gerda and said, "They shan't kill you unless I get angry with you.
THE SNOW QUEEN

Cæsium-137 is accumulated in muscle tissue, just like potassium - its chemical congener. The concentration in the human body is conveniently measured together with potassium in a whole-body counter. Due to the relatively short biological halflife of ^{137}Cs in the human body (approx. 110 days (Ek66)), the levels in the diet and in the body attain equilibrium relatively rapidly. Cæsium-137 was first detected in humans in 1956 (Mi56). The first more comprehensive assessments were carried out at Los Alamos, USA (An57), and at Harwell, UK (Ru60). At present, routine measurements are made in France, West Germany, Sweden, Switzerland, Finland, USSR, Japan, Argentina and Australia (Un77). The Danish study began in 1963 and has been performed on a control group of Risø staff, numbering approx. 20 adults, who were not occupationally exposed to ^{137}Cs. The group has been measured each year in the spring, late summer and winter. In adults (> 20 years), ^{137}Cs concentrations (often given as pCi ^{137}Cs $(g\ K)^{-1}$) are with approximation independent of age (On62), but in children and adolescents the levels may differ from those found in adults due to a shorter biological halflife of ^{137}Cs in the younger age groups (Be67, Bo69).

4.5.1. Variation with time, sex and individual

The maximum level of ^{137}Cs in the Risø control group occurred in 1964 (fig. 4.5.1.) with a mean concentration of 162 pCi ^{137}Cs $(g\ K)^{-1}$ corresponding to a wholebody content in standard man ($\sim$ 140 g K) of approx. 23 nCi ^{137}Cs (RRD64). Since then

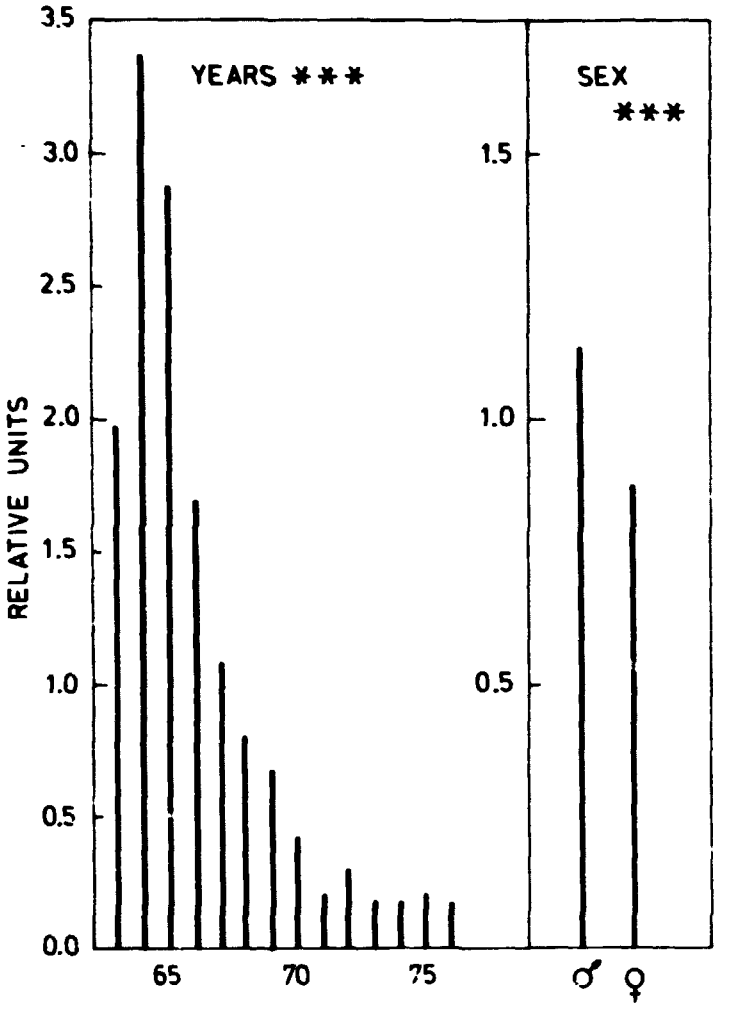

Fig. 4.5.1. The variation of pCi ^{137}Cs (g K)$^{-1}$ in adult human bodies in Zealand (cf. Table A.1.4.5.) during 1963-1976. The bars show the levels relative to the grand mean 58 pCi (g K)$^{-1}$ (= 1 on the relative scales).

concentrations have decreased and by the middle of the seventies the mean level was approx. 10 pCi ^{137}Cs (g K)$^{-1}$. The variation among years was thus highly significant (table B.4.5.1.), the variability was 1.13 (table B.4.5.2.) and not significantly different from that of ^{137}Cs in total diet (B.4.2.3.). An anova of all data with year and sex as main factors showed a significant variation between the two sexes. The ^{137}Cs mean concentration (pCi ^{137}Cs (g K)$^{-1}$) was 1.3 times higher in males than in females (fig.4.5.1.). This intersexual difference became even more pronounced if the pCi ^{137}Cs kg^{-1} body weight figures were considered, because the potassium concentrations in

females are lower than in males. UNSCEAR (Un72) reports that the pCi ^{137}Cs (g K)$^{-1}$ in women is 20-30% lower than in men. The phenomenon is thus general, and it may be due to a difference in metabolism between males and females. It is thus known that the basal metabolic rate in women is lower than that of men of comparable size (Ek66). The biological halftime of ^{137}Cs is probably longer for males than for females (Be67, Bo69). The phenomenon could, however, also be influenced by differences in composition of the diet between the two sexes. It has been demonstrated (RRD64) that vegetarians contain lower ^{137}Cs levels than other adults. If the male diet on the average contained more meat and bread than the female diet, which may be enriched with respect to vegetables and fruit, the ^{137}Cs (g K)$^{-1}$ ratio of the male diet would be higher than that of the female. The variability between sexes increased with time (table B.4.5.2.), the difference between males and females has thus become more pronounced in later years. Reference groups in West Germany (De76) and Switzerland (Hu74) showed decreasing ratios between men and women in the same period. As there seems no evident explanation for a decreasing or increasing variability between sexes, the observation may be fortuitous.

4.5.2. Relations and prediction models

The radioecological sensitivity of the human body to ^{137}Cs contamination was calculated from the prediction models in table C.4.5.1. The sensitivity of the male body to ^{137}Cs contamination was 1.3 times that of the female. The models indicated that there was no strong influence of old deposited ^{137}Cs, and thus there were no indications of a ^{137}Cs pool in the body with a slow turnover rate as earlier suggested (X). This agreed with the observations of HARDY (Ha74a), which showed that ^{137}Cs was barely measurable in specimens of calcified bone tissue. From the models for ^{137}Cs in diet (table C.4.2.2 Nos.4 and 5) and the whole body models, the transfer factor from diet to body of ^{137}Cs was estimated; P_{34} ($= \frac{P_{234}}{P_{23}}$) became 2.85 pCi ^{137}Cs (g K)$^{-1}_{body}$ per pCi ^{137}Cs (g K)$^{-1}_{diet}$, which agreed with earlier estimates (X, Un77). As the best available estimate for the radioecological sensitivity of the Danish human body to ^{137}Cs concentration from fallout, the mean of the two estimates Nos. 1 and 2 in

table C.4.5.1. was applied, i.e. 11.5 ± 0.3 (1 SE) pCi ^{137}Cs $(g\ K)^{-1}$ • y per mCi ^{137}Cs km^{-2}. In standard man (Ic59) the potassium content is 140 g, hence 1 mCi ^{137}Cs km^{-2} corresponded to 1.6 nCi ^{137}Cs • y $(cap)^{-1}$. In the case of ^{90}Sr, where standard man contains 1,100 g Ca, the infinite body-burden time integral of 1 mCi ^{90}Sr km^{-2} was estimated at 0.90 nCi ^{90}Sr • y $(cap)^{-1}$ (cf.4.3.2.). Thus the radioecological sensitivity of the human body to ^{137}Cs was 1.8 times that of ^{90}Sr, i.e., by chance nearly the same factor as found for the total diet intakes (4.2.2.).

4.6. Human milk

Saa mødte han en Amme med et lille Barn. "Hør du Tyrke-Amme!" sagde han, "hvad er det for et stort Slot her tæt ved Byen, Vinduerne sidde saa høit!"
DEN FLYVENDE KOFFERT

Then he met a nurse with a baby. "I say, you Turk-nanny", he began, "what's this great castle here, close to the town, with the high windows?"
THE FLYING TRUNK

Human milk is a unique link in the human food chain, because it is both produced and utilized by the human organism. Measurements of the radioactive contamination of human milk have been scarce, partly because it is difficult to obtain samples, partly because breastfeeding, at least in the western world, is a minor source of radioactive contamination for infants, as mothers' milk is often substituted by cows' milk products. The first information on radionuclides in human milk originated from measurements of a few individual samples collected in the UK in 1957 in connection with the Windscale accident (Ma58a). A systematic study of ^{90}Sr in human milk was carried out in the USA in 1959 (Lo60), and the first study of the secretion of ^{137}Cs in human milk was performed in 1962 (XIII). Danish studies of fallout nuclides in human milk continued until 1969, when levels were so low that it had become difficult to make reasonably accurate measurements on the samples obtainable. However, the Danish study of ^{90}Sr and ^{137}Cs in human milk is the only study covering several years mentioned in the literature.

4.6.1. Variation with time

In analogy with cows' milk (3.2.), human milk was investigated within the so-called milk year, i.e. the period from May in a

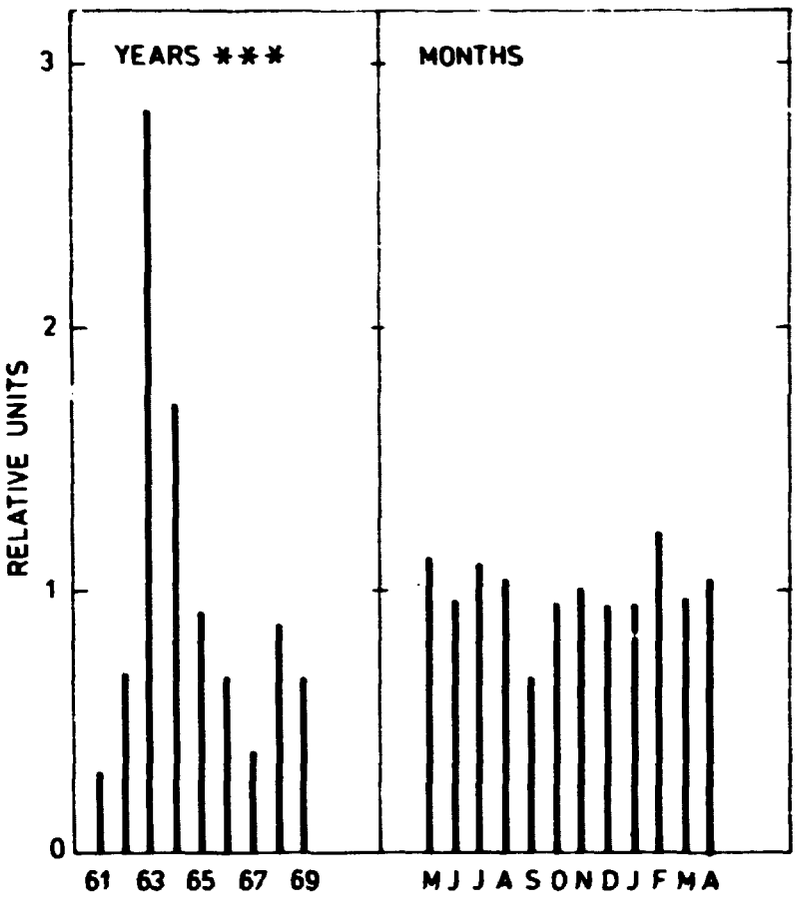

Fig. 4.6.1.1. The variation of pCi ^{90}Sr (g Ca)$^{-1}$ in human milk collected in 1961-1969 in Zealand (cf. Table A.1.4.6.). The bars show the levels relative to the grand mean 2.2 pCi (g Ca)$^{-1}$ (= 1 on the relative scale). The years were the "milk years" (cf. the text).

given year until April in the following year. The relative mean levels shown in figs. 4.6.1.1. and 4.6.1.2. were calculated from VAR-3 (table B.4.6.1.), assuming no interaction between milk-year and month. The samples contained 0.27 $\pm$ 0.10 Ca l^{-1} (1 SD) (mean of 28 determinations) and 0.58 $\pm$ 0.12 g K l^{-1} (135 determinations), which was in agreement with observations in the literature (Sp56, Lo60, Str65, Ca70), considering that the Danish human milk samples were all mature milk, i.e. secreted more than 10 days after parturition. The ^{90}Sr concentrations showed a highly significant variation among years but no variation among months. The variability (table B.4.6.2.) among years was 0.59, i.e. compatible with that observed for ^{90}Sr in cows' milk (3.2) and in total diet (4.2). In the case of ^{137}Cs the variability was 1.03, also in accordance with the observations for milk and total diet. The variability of ^{137}Cs

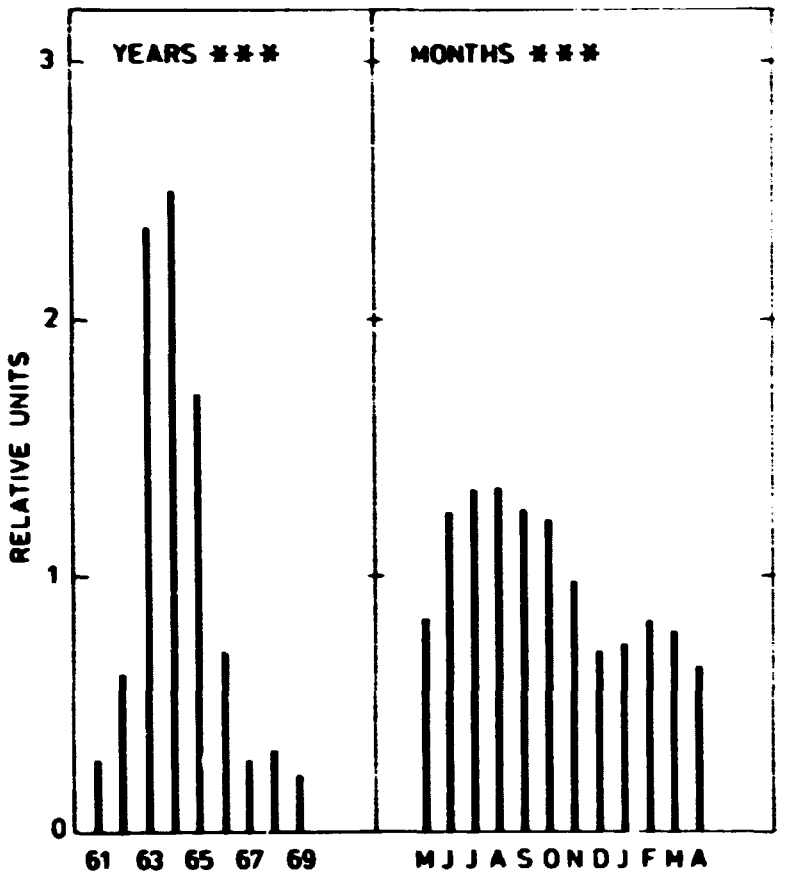

Fig. 4.6.1.2. The variation of pCi ^{137}Cs (g K)$^{-1}$ in human milk collected in 1961-1969 in Zealand (cf. Table A.1.4.6.). The bars show the levels relative to the grand mean 43 pCi (g K)$^{-1}$ (= 1 on the relative scale). The years were "milk years" (cf. the text).

among months was 0.18, higher than for ^{90}Sr ($CV_{p_{month}}$ = 0.11), but lower than the variability among months for ^{137}Cs in cows' milk, indicating that the other constituents of the human diet (especially bread) varied less throughout the year than cows' milk.

4.6.2. Relations and prediction models

During the period of human milk sampling, the diet intakes of ^{90}Sr and ^{137}Cs were determined on four occasions. The milk donors prepared double portions of everything they consumed during seven days, and their milk was sampled and analyzed in the same period. These experiments showed that 33% ± 13% (1 SD) of the daily intake of ^{137}Cs were secreted per litre of milk and 2.3% ± 1.05% of the daily ^{90}Sr intake. The percentages could also be estimated from the total diet measurements carried

out on samples collected in Zealand and Copenhagen (RRD69, RRD59-76). In this case the percentages became 22% ± 9% for ^{137}Cs and 2.6% ± 1.05% for ^{90}Sr. LOUGH et al. (Lo60) found 1.5% ± 0.42% for ^{90}Sr, and CALAPAJ et al. (Ca70) found 1.6% ± 1.4% for ^{90}Sr and 75% ± 37% for ^{137}Cs. In an experimental study of ^{137}Cs secretion in human milk (XIII), 10% per cent of a single acute intake were secreted with the milk within two weeks after intake. The secretion of ^{137}Cs followed a two-component exponential curve with halftimes of approx. 2 and 7 days within the period of observation. The above higher percentages for chronic intakes, however, indicated that the secretion of ^{137}Cs in human milk may rather follow a three-component exponential curve with a final halflife equal to the biological halftime of ^{137}Cs in the human body. The estimated ^{137}Cs content in milk from "normal fallout" contaminated diet may thus have been too high in the experiment (cf. table 3 in (XIII)).

On six occasions (RRD64, RRD65) milk donors were measured during lactation for wholebody ^{137}Cs in 1964-1965. The observed mean ratio between pCi ^{137}Cs (g K)$^{-1}$ in milk and body was 0.53 ± 0.08 (1 SD). BENGTSSON et al. (Be64b) found experimentally, for a single individual, a ratio of 0.2 between the specific (per kg wet weight) concentrations of ^{137}Cs in milk and the whole body. Assuming that milk and body contained respectively 0.6 g K kg^{-1} and 1.8 ± 0.5 (1 SD) g K kg^{-1}, the agreement between the two observed ratios was satisfactory. Instead of calculating a special prediction model for ^{137}Cs in human milk, it was more expedient to use the models for ^{137}Cs in women from Zealand (table C.4.5.1., Nos. 5 and 6) multiplied by 0.53 (fig. 4.6.2.1). Hereby the radioecological sensitivity of human milk to ^{137}Cs fallout was estimated at 5.3 pCi ^{137}Cs (g K)$^{-1}$ y per mCi ^{137}Cs km^{-2} or 3.1 pCi ^{137}Cs l^{-1} y per mCi ^{137}Cs km^{-2}. A woman with a daily milk production of 1 litre may thus in a year have secreted 1.1 nCi ^{137}Cs with her milk, or $\frac{1.1}{5.6}$ = 20% of her ^{137}Cs intake with the diet (cf.4.2.2). This estimate was compatible with the above observations. As pCi ^{137}Cs (g K)$^{-1}$ y is higher in Danish human milk than in Danish cow milk, higher time-integrated ^{137}Cs body concentrations were to be expected in Danish babies fed on human milk instead of cows' milk. RUNDO (Ru70) found in a study performed in the UK in 1963-1964, using both breast- and bottle-fed babies, that babies

fed on human milk showed lower body concentrations than babies fed on cows' milk. However, in RUNDO's study the pCi ^{137}Cs $(g\ K)^{-1}$ levels were approx. 20% higher in cows' milk than in human milk.

In the case of ^{90}Sr, mothers' milk shows nearly the same pCi ^{90}Sr $(g\ Ca)^{-1}$ ratio as the bone of newborn children, i.e. the discrimination against Sr relative to Ca was nearly the same via the placenta as via the mammary gland (Co61a). If the half-year periods between January 1964 and December 1969 are considered, the mean observed ratio between human milk and bone of newborn children became 1.09 $\pm$ 0.14 (1 SE) (11 periods), and the correlation coefficient between milk and bones 0.702, i.e. significant. Hence the prediction model for ^{90}Sr in bone of newborn children was applied as a model for ^{90}Sr in human milk. Hereby the radioecological sensitivity of human milk to ^{90}Sr fallout became 1 pCi ^{90}Sr $(g\ Ca)^{-1} \cdot y$ per mCi $^{90}Sr\ km^{-2}$ (table C.4.3.1., Nos. 1 and 2), or 0.3 pCi $^{90}Sr\ l^{-1}\ y$ per mCi $^{90}Sr\ km^{-2}$. In analogy with ^{137}Cs, the % of secretion with milk of the total diet ^{90}Sr intake was $\frac{0.1}{3.0} = 3.6\%$, i.e. in reasonable agreement with the measurements.

4.7. Conclusions

4.7.1. General

Man is an omnivore and the total human diet thus contains vegetable as well as animal products. In general, the first category contains the important ^{90}Sr donors, whereas animal products are the main source of ^{137}Cs. However, the two important diet groups, milk and cereals, are both pronounced ^{90}Sr and ^{137}Cs donors despite their origin. Faroese and Greenlandic diets are characterized by a relatively high ^{137}Cs content; this is mainly because local produce from these areas is of animal origin, while most vegetable products are imported. The Danish, Faroese and Greenlandic populations show a higher radioecological sensitivity to environmental contamination with ^{137}Cs than with ^{90}Sr. The calculated wholebody doses from internal contamination originating from nuclear weapons fallout,

based on the ICRP weighting factors for the various organ doses, were higher for ^{137}Cs than for ^{90}Sr. If furthermore the contribution from external radiation from deposited ^{137}Cs was considered, ^{137}Cs became definitely the most hazardous of the two radionuclides.

4.7.2. Total diet

The variability among years of ^{90}Sr in the Danish total diet ($CV_{p\ years} = 0.6$) was lower than that in milk and bread, but higher than that in vegetables. The variability of ^{137}Cs ($CV_{p\ years} = 1.3$) was close to that in meat but lower than that in milk and bread and higher than that in fish.

The local variability of ^{90}Sr and ^{137}Cs in the Danish total diet was nearly the same for the two nuclides ($CV_{p\ location} = 0.13$). Milk showed a higher local variability of these nuclides than total diet, white bread a lower, but rye bread and vegetables nearly the same variability.

The ratios between the $^{90}Sr/Ca$ quotients in the total diet and in milk showed significant variations among years as well as among locations, and so did $^{137}Cs/K$ quotients. Milk measurements were thus no possible substitute for total diet measurements.

The radioecological sensitivities of the Danish total diet were 5.1 pCi ^{90}Sr $(g\ Ca)^{-1}$ y per mCi ^{90}Sr km^{-2} and 4.4 pCi ^{137}Cs $(g\ K)^{-1}$ y per mCi ^{137}Cs km^{-2}. The sensitivity to ^{137}Cs contamination was 1.08 times higher in Jutland than in the Islands; but the two areas of the country did not differ with respect to sensitivity to ^{90}Sr. The infinite time-integrated intakes with the Danish total diet originating from fallout were 0.23 µCi ^{90}Sr $(cap)^{-1}$ ($8.5 \cdot 10^3$ Bq) and 0.66 µCi ^{137}Cs $(cap)^{-1}$ ($2.4 \cdot 10^4$ Bq). Grain products contributed approx. 40% of the radioecological sensitivity of the Danish total diet for ^{90}Sr as well as for ^{137}Cs, milk 30% and 17%, respectively, vegetables and fruit 26% and 10%, and meat and eggs 2% and 32%.

In the case of exclusive indirect contamination of the total Danish diet, milk products, cereals and vegetables would each

contribute about one third to the total ^{90}Sr intake from the diet. The $^{90}Sr/Ca$ quotient of the diet would in the case of indirect contamination be 25% of the quotient in the ploughing layer of Danish soil, provided that all calcium in the diet was derived from crops; if calcium in creta praeparata and drinking water was included, the percentage would be halved.

The Faroese total diet showed slightly less radioecological sensitivity to ^{90}Sr contamination than the Danish, but in the case of ^{137}Cs the sensitivity of the Faroese diet was three times higher than the Danish, due to its content of Faroese milk, potatoes and mutton. The infinite time-integrated intakes with the Faroese total diet originating from nuclear weapons fallout were 0.34 μCi ^{90}Sr $(cap)^{-1}$ ($1.3 \cdot 10^4$ Bq) and 3.7 μCi ^{137}Cs $(cap)^{-1}$ ($1.4 \cdot 10^5$ Bq).

The radioecological sensitivity to ^{90}Sr contamination of the Greenlandic total diet was similar to that of the Danish, while the sensitivity to ^{137}Cs was twice as high, because of the consumption of Greenlandic mutton and reindeer. The infinite time-integrated intakes of the two nuclides with the Greenlandic total diet were 0.17 μCi ^{90}Sr $(cap)^{-1}$ ($6.5 \cdot 10^3$ Bq) and 1.0 μCi ^{137}Cs $(cap)^{-1}$ ($3.6 \cdot 10^4$ Bq).

4.7.3. Human bone

The variability among years of the levels of ^{90}Sr in human bone ($CV_{p\ years} = 0.4$) was lower than that of ^{90}Sr in the diet as a result of the relatively long residence time of ^{90}Sr in bone. The time variabilities of the various age groups were not much different, but infants and teenagers showed higher values than the average because of the rapider turn-over rates of Sr-Ca in these age groups.

The local variability ($CV_{p\ locations} = 0.1$) of ^{90}Sr in human bone was also lower than that of the diet because people do not always remain in the same part of the country for life. Adult bone from East Jutland contained 10% higher levels than adult bone from Zealand. The ^{90}Sr levels in bone showed no significant differences between the two sexes.

The observed ratio between the $^{90}Sr/Ca$ quotients in adult vertebrae and diet was 0.16. The turnover rate of ^{90}Sr in adult vertebrae ($\geq$ 29 yr) was estimated at 0.16 y^{-1}, corresponding to an effective halflife of 3.8 years. The observed ratio between the $^{90}Sr/Ca$ quotients in bone of newborn infants and the $^{90}Sr/Ca$ quotients in the adult diet increased from 0.1 in the first part of the sixties to approx. 0.2 in the seventies, suggesting the transfer of ^{90}Sr from maternal bone to the fetal skeleton.

The radioecological sensitivity of adult human vertebrae in Denmark was estimated at 0.82 pCi ^{90}Sr $(g\ Ca)^{-1}$ y per mCi ^{90}Sr km^{-2}. Infants (4-12 months old) showed the highest sensitivity of all age groups, namely 1.4. The radioecolcgical sensitivity decreased with increasing age from infants to adults. The creta praeparata in the diet has reduced the ^{90}Sr concentration in Danish bone.

4.7.4. Human teeth

After formation during pregnancy and the first year of life, the shed deciduous tooth crown does not exchange significant amounts of ^{90}Sr with its surroundings. The ^{90}Sr concentration in shed deciduous teeth from a tooth donor born in the year (i-1) corresponded to that of his bone when he was approx. 1 year old in the year (i).

Teeth from Jutland showed 20% higher ^{90}Sr levels than teeth from the Islands. Greenlandic teeth contained concentrations similar to the Danish, but Faroese teeth had higher ^{90}Sr levels. The variability among years ($CV_{p\ years}$ = 1.1; 1950-1969) was the same for Denmark, the Faroes and Greenland. The radioecological sensitivity of Faroese teeth to ^{90}Sr contamination was 1.2 times that of the Danish. From this value, the infant Faroese bone levels were estimated at 1.2 • 2.08 = 2.5 times the Danish levels. The Greenlandic tooth levels suggested that the infant bone levels were similar to those observed in Denmark.

The ^{90}Sr concentrations in teeth suggested that the estimated fallout levels for the first years of nuclear testing prior to 1959 are not seriously encumbered by error.

4.7.5. Human body

The variability among years of ^{137}Cs in the human body ($CV_{p\ years}$ = 1.1) was similar to that of the total diet. Males showed 1.3 times higher pCi ^{137}Cs $(g\ K)^{-1}$ levels than females.

The observed ratio between human body and total diet $^{137}Cs/K$ quotients was 2.85. The radioecological sensitivity of the human body was 11.5 pCi ^{137}Cs $(g\ K)^{-1}$ y (1.6 nCi ^{137}Cs $(cap)^{-1}$ y) per mCi ^{137}Cs km^{-2}. The infinite time exposure integral from a total deposit of 117 mCi ^{137}Cs km^{-2} was 0.19 µCi ^{137}Cs $(cap)^{-1}$ y ($7.0 \cdot 10^3$ Bq), compared to a ^{90}Sr time integral of 0.07 µCi ^{90}Sr $(cap)^{-1}$ y ($2.4 \cdot 10^3$ Bq) from 73 mCi ^{90}Sr km^{-2}.

4.7.6. Human milk

The variabilities among years of ^{90}Sr ($CV_{p\ years}$ = 0.5) and ^{137}Cs ($CV_{p\ years}$ = 1.0) in Danish human milk were similar to those of cows' milk. The variability of human milk among months was higher for ^{137}Cs ($CV_{p\ month}$ = 0.2) than for ^{90}Sr ($CV_{p\ months}$ = 0.1), but the difference was less pronounced than for cows' milk.

Of the daily diet intakes, 33 ± 13% (1 SD) of the ^{137}Cs and 2.3 ± 1.05% of the ^{90}Sr was secreted per liter human milk; these percentages were approx. 50 and 10 times higher than the corresponding ones for cows' milk. Even if the higher milk production of the cow was taken into acount, the human female still secreted a larger proportion of the diet intake of ^{137}Cs with her milk than the cow.

The radioecological sensitivities of Danish human milk were estimated at 5.3 pCi ^{137}Cs $(g\ K)^{-1}$ y (or 3.1 pCi $^{137}Cs\ l^{-1}$ y) per mCi ^{137}Cs km^{-2} and 1 pCi ^{90}Sr $(g\ Ca)^{-1}$ y (or 0.3 pCi $^{90}Sr\ l^{-1}$ y) per mCi ^{90}Sr km^{-2}. Hence a Danish baby living solely on human milk would have a higher ^{137}Cs body burden, but

a lower ^{90}Sr content than a baby living on cows' milk only. A Faroese baby would receive more ^{137}Cs as well as ^{90}Sr from cows' milk than from mother's milk. A Greenlandic baby gets a higher ^{137}Cs body content if it is fed on mother's milk instead of Danish cows' milk.

4.7.7. Dose estimates and risk evaluations (cf.table 4.7.7.)

In the case of Denmark doses to the population from ^{90}Sr were estimated from the radioecological sensitivity of adult human bone, whereas the corresponding estimates for the Faroes and Greenland were based on the sensitivities of the respective diets (tables 4.2.4. and 4.2.5.) and on the transfer factor P_{34} = 0.16 pCi ^{90}Sr (g Ca)$^{-1}$ y in bone per pCi ^{90}Sr (g Ca)$^{-1}$ y in diet estimated from Danish diet and bone measurements. The integral deposits of ^{90}Sr in the three areas were 73 mCi km^{-2} in Denmark, 152 mCi km^{-2} in the Faroes, and 55 mCi km^{-2} in Greenland (west coast), and the transfer factors: P_{45} (tissue to dose) were, according to UNSCEAR, (Un77) for bone marrow: 1.4 mrad per pCi ^{90}Sr (g Ca)$^{-1}$ y and for bone lining cells: 1.9 mrad per pCi ^{90}Sr (g Ca)$^{-1}$ y.

The internal doses from ^{137}Cs were estimated from the radioecological sensitivities of total diet (tables 4.2.2., 4.2.4. and 4.2.5.) and from the transfer factor P_{34} = 2.85 pCi ^{137}Cs (g K)$^{-1}$ y in soft tissue per pCi ^{137}Cs (g K)$^{-1}$ y in diet estimated from Danish diet and wholebody measurements. The integral deposits of ^{137}Cs were 1.6 times those of ^{90}Sr, i.e. 117 mCi km^{-2} in Denmark, 243 mCi km^{-2} in the Faroes and 88 mCi km^{-2} in Greenland, and the transfer factor P_{45} from tissue to dose to wholebody was: $1.8 \cdot 10^{-2}$ mrad per pCi ^{137}Cs (g K)$^{-1}$ y (Un77).

The external dose factor for ^{137}Cs was 1.44 mrad per mCi ^{137}Cs km^{-2} (Un77); 0.32 (see the table) was a factor estimated by UNSCEAR which, among other things, took the shielding effect of buildings into consideration. This factor was considered constant for the various radionuclides contributing to external dose.

Table 4.7.7. Dose commitments from nuclear weapons debris through 1975.

	Denmark (5×10^6 inhabitants)	Faroes (4×10^4 inhabitants)	Greenland' (5×10^4 inhabitants)
Bone marrow dose from ^{90}Sr	1.4×73×0.82=84 mrad (0.84 mGy)	1.4×152×0.16×5.40= 184 mrad (1.84 mGy)	1.4×55×0.16×7.16= 88 mrad (0.88 mGy)
Dose to bone lining cells from ^{90}Sr	1.9×73×0.82=114 mrad (1.14 mGy)	1.9×152×0.16×5.40= 250 mrad (2.50 mGy)	1.9×55×0.16×7.16= 120 mrad (1.20 mGy)
Whole-body dose from body ^{137}Cs	0.018×117×4.4×2.85= 26 mrad (0.26 mGy)	0.018×243×13.05×2.85= 163 mrad (1.63 mGy)	0.018×88×9.96×2.85= 45 mrad (0.45 mgy)
Whole-body dose from external ^{137}Cs	1.44×0.32×117= 54 mrad (0.54 mGy)	1.44×0.32×243= 112 mrad (1.12 mGy)	1.44×0.32×88= 41 mrad (0.41 mGy)
Lung dose from ^{106}Ru	1×27=27 mrad (0.27 mGy)	27 mrad (0.27 mGy)	27 mrad (0.27 mGy)
Lung dose from ^{144}Ce	2.3×22=51 mrad (0.51 mGy)	51 mrad (0.51 mGy)	51 mrad (0.51 mGy)
Total body (weighted) $^{239,240}Pu$ through 2000	0.1 mrad (1 μGy)	0.1 mrad (1 μGy)	0.1 mrad (1 μGy)
External dose from ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{140}Ba, ^{141}Ce, ^{144}Ce	28+2+12+3+0+2= 47 mrad (0.47 mGy)	47×2.08=98 mrad (0.98 mGy)	47×0.75=35 mrad (0.35 mGy)
Total body dose from ^{3}H	2 mrad (0.02 mGy)	2 mrad (0.02 mGy)	2 mrad (0.02 mGy)
Total body (weighted) dose from ^{14}C through 2000	8 mrad (0.08 mGy)	8 mrad (0.08 mGy)	8 mrad (0.08 mGy)
Tissue-weighted [a] whole-body dose equivalent commitment	1.62 mSv (162 mrem)	4.24 mSv (424 mrem)	1.57 mSv (157 mrem)
Collective dose [b] equivalent commitment	8100 man·Sv	170 man·Sv	78 man·Sv

As the mortality risk factor for radiation-induced cancers from uniform whole-body irradiation is about $10^{-2} \cdot Sv^{-1}$ (Ic 77), the estimated number of deaths from such cancers from a collective dose of 8348 man·Sv is 83. The expected number of serious cases of ill-health in future generations due to hereditary detriment following irradiation was also of the order of 100 (Ic 77). Cesium-137 was responsible for 50% of the collective dose equivalent commitment in Denmark, for 65% in the Faroes and for 55% in Greenland.

The total radioecological sensitivity of the Faroese and Greenlandic populations was approx. 30% higher than that of the Danish (Faroes: $\frac{4.24}{2.08\times1.62} = 1.26$; Greenland: $\frac{1.57}{0.75\times1.62} = 1.29$).

[a] also called per caput effective dose equivalent commitment.

[b] here identical with collective effective dose equivalent commitment (D.4.7.7).

The integrated air concentrations of ^{106}Ru and ^{144}Ce were taken from table 1.2.5. The dose factors: 27 and 22 mrad, respectively, to lung tissue per pCi m^{-3} y were those estimated by UNSCEAR (Un77).

The tissue doses from $^{239,240}Pu$ were estimated in 1.7.2. The weighted wholebody dose was calculated by means of the ICRP weighting factors (Ic77) (gonads: 0.25, breast: 0.15, red bone marrow: 0.12, lung: 0.12, thyroid: 0.03, bone surfaces: 0.03, remainder: 0.30 (distributed on five organs, each attributed a weighting factor of 0.06)). (cf. also D.4.7.7).

The integral deposits of ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce and ^{144}Ce were estimated from the integral air concentrations of these radionuclides (table 1.2.5.) assuming a wash-out factor of 1 pCi l^{-1} per fCi m^{-3}, i.e. equal to that of ^{90}Sr (1.3.2.), and a rainfall of 628 mm y^{-1} in Denmark. Hence the integral Danish deposits became: 1005 mCi ^{95}Zr km^{-2}, 440 mCi ^{103}Ru km^{-2}, 628 mCi ^{106}Ru km^{-2}, 276 mCi ^{141}Ce km^{-2} and 1,444 mCi ^{144}Ce km^{-2}. The dose conversion factors for the five radionuclides were 0.087, 0.011, 0.060, 0.0014 and 0.0045 mrad per mCi km^{-2}, respectively (Un77), as in the case of ^{137}Cs a shielding factor of 0.32 was furthermore applied. Danish data on ^{140}Ba was not available, therefore the UNSCEAR estimate for the northern temperate zone was applied instead. The Faroese and Greenlandic doses were obtained from the Danish ones by multiplying by 2.08 and 0.75, respectively, i.e. the ratios between ^{90}Sr fallout in the respective areas and in Denmark.

The doses from ^{3}H and ^{14}C were those estimated by UNSCEAR (Un77) for the northern temperate zone. The doses from ^{14}C were transformed into wholebody doses by means of the ICRP weighting factors. Contributions from other radionuclides were not taken into account as their doses were considered to be less than 1% of the total dose from the nuclides considered above.

The tissue-weighted wholebody dose equivalent was calculated from the ICRP weighting factors assuming the quality factor to be 20 for the plutonium dose and 1 for the other nuclides in table 4.7.7.

4.7.8. Concluding remarks on radioecological sensitivity and variability

Radioecological sensitivity and variability were applied to quantify the response of environmental samples to radioactive contamination from nuclear weapons debris. Three areas were studied: Denmark, the Faroe Islands and Greenland. As appears from table 4.7.8, the radioecological sensitivities found in various types of sample from these areas displayed a wide range of values; thus, for the same nuclide and sample type, the radioecological sensitivity varied between two locations by, e.g., a factor of 20 for ^{137}Cs in Faroese and Danish grass. Between nuclides in the same type of sample, e.g., Faroese mutton, the sensitivities to ^{137}Cs and ^{90}Sr contamination differed by a factor of 70. Between different species of sample, Greenland lichen showed, e.g., nearly 14000 times higher sensitivity to ^{137}Cs contamination than did Danish cabbage.

Table 4.7.8 may also be used to estimate the transfer factors from air to the various environmental samples; e.g. if the annual mean air concentration at steady state was 1 fCi ^{137}Cs kg^{-1}, then the Greenland lichen ^{137}Cs concentration would be 14 nCi kg^{-1}, Faroese milk 0.1 nCi kg^{-1} and Danish cabbage 1 pCi kg^{-1}.

In this study radioecological sensitivity has been defined as the transfer factor from deposition to the environmental sample. However, on various occasions the environmental radioactivity levels indicated some independence of the activity deposited with rainfall, i.e. radioecological sensitivity depended upon precipitation in such a way that increased rainfall was tantamount to a lower sensitivity of the vegetation sample due to wash-off of the adsorbed debris. In earlier studies (Aa66a, Aa66c, X) prediction models applying air concentrations instead of deposition with rainfall have been suggested. In the case of prevailing direct contamination, such models were generally successful. Radioecological sensitivity could thus have been based on air concentrations instead of deposition. However, as precipitation measurements are necessary to assess the indirect contamination from accumulated deposits, the present method of

Table 4.7.8. Infinite time integral concentrations in various environmental samples resulting from a deposition of 1 mCi km^{-2} of ^{90}Sr or ^{137}Cs, respectively.

	Denmark				The Faroes				West Greenland			
	pCi $^{90}Sr \cdot y$		pCi $^{137}Cs \cdot y$		pCi $^{90}Sr \cdot y$		pCi $^{137}Cs \cdot y$		pCi $^{90}Sr \cdot y$		pCi $^{137}Cs \cdot y$	
	kg^{-1}	$(g\ Ca)^{-1}$	kg^{-1}	$(g\ K)^{-1}$	kg^{-1}	$(g\ Ca)^{-1}$	kg^{-1}	$(g\ K)^{-1}$	kg^{-1}	$(g\ Ca)^{-1}$	kg^{-1}	$(g\ K)^{-1}$
Air[a]	1.3×10^{-3}	-	1.3×10^{-3}	-	0.6×10^{-3}	-	0.6×10^{-3}	-	1.3×10^{-3}	-	1.3×10^{-3}	-
Rain[b]	1.6	-	1.6	-	0.7	-	0.7	-	1.6	-	1.6	-
Soil[c]	46	17	46	3	152	-	152	80	60	-	60	3
Sea water	0.35	1.9	0.23	1.4	0.03	0.08	0.03	0.08	0.23	0.7	0.23	0.8
Drinking water[d]	7×10^{-3}	9×10^{-2}	-	-	0.1	-	-	-	0.5	-	-	-
Rye	32	79	46	11	-	-	-	-	-	-	-	-
Wheat	24	58	28	8	-	-	-	-	-	-	-	-
Grass	38	31	24	5	250	200	500	75	550	450	350	80
Cabbage	8	17	1.3	0.5	-	-	-	-	-	-	-	-
Potato	2.6	52	3.8	1.0	2.8	100	44	14	-	-	-	-
Lichen	-	-	-	-	-	-	-	-	1.6×10^{3}	-	18×10^{3}	-
Sea plants	110	11	-	-	12	1.2	-	-	60	6	-	-
Rye bread[e]	23	8	38	11	7	4	12	4	-	-	-	-
White bread[e]	4	2	10	7	2	2	4	3	-	-	-	-
Milk	3.9	3.3	5.5	3.4	10	9	60	35	-	-	-	-
Meat[f]	1.4	14	28	8	3	30	200	80	5	50	135	350
Cod	0.09	0.9	20	6	0.06	0.6	1.7	0.5	-	-	10	3
Total diet	3	5.1	5	4.4	2.1	5.4	14	13	3.3	7	11	10
Human bone[g,h]	300	0.8	-	-	-	(0.9)	-	-	-	(1.2)	-	-
Human body[h]	-	-	23	11.5	-	-	-	(40)	-	-	-	(30)
Human milk[h]	0.3	1	3	5	-	(1.1)	-	(18)	-	(1.4)	-	(14)

a) Estimated from the rainwater concentrations assuming a scavenging factor of 0.99 pCi l^{-1} rain per fCi m^{-3} air and a specific air density of 1.23 kg m^{-3}.

b) Estimated from an annual mean precipitation of 628 mm in Denmark, 1500 mm in the Faroes, and 630 mm in West Greenland.

c) Estimated from an effective half-life of ^{90}Sr and ^{137}Cs in the 0-50 cm soil layer of 22.5 y (1.6.1) The mean volume weight of soil was 1.4 g cm^{-3} in Denmark (RRD 75), 0.43 g cm^{-3} in the Faroes, and 1.1 g cm^{-3} in West Greenland (cf. Table D.1.6.1.2).

d) The figures used for Denmark were those calculated for ground water.

e) The radioecological sensitivities of Faroese bread were derived from the sensitivities calculated in Table C.2.3.1. divided by 2.08 (cf. 4.2.4.).

f) Beef in Denmark, and mutton in the Faroes and Greenland.

g) Adult human bone.

h) The Faroese and Greenlandic figures were estimated from the respective diet levels and from the Danish transfer factors.

estimating the radioecological sensitivity was preferred. It should, however, be recalled that the radioecological sensitivity may not be althogether independent of precipitation, (cf. also D.4.7.8).

The variability with time of the concentration of a radionuclide in an environmental sample is a useful way to assess the route of contamination of the sample. A low variability among years generally suggested strong dependence upon root uptake (indirect contamination), or of a long residence time of the debris in a directly contaminated sample such as observed in, e.g., lichen, while a high variability was indicative of prevailing direct contamination combined with a short residence time of the debris in the sample. In the case of nearly constant fallout rate, the annual variabilities of direct and indirect contamination approach each other. Hence it should be recalled that the interpretation of variability with regard to routes of contamination is linked to the variability of fallout rate and accumulated deposit.

The local variability of radionuclide levels is a measure of environmental inhomogenity with respect to radioactive contamination. The local variability depends upon fallout and on the radioecological sensitivity of the area considered. A low local variability is tantamount to the area being relatively homogeneous to radioactive contamination, implying that the number of sample locations in such an area can be relatively low. The local variability may vary with time. This could arise from variations in the relative contributions from direct and indirect contamination, from alterations in agricultural practice, or from the transfer of foodstuffs from other areas into the area in question. Such variations of the local variability imply current revisions of the environmental surveillance programmes.

In conclusion, radioecological sensitivity and variability have shown to be useful quantities for characterizing the radioecological properties of environmental samples contaminated by global fallout from nuclear weapons testing.

5. GENERAL DISCUSSION

5.1. Introduction

The preceding four chapters have been a presentation of data, and we have seen how proper treatment of the data could characterize the radioecological properties of various environments.

Chapter five aims to show various applications of the results obtained. The chapter is more speculative and less substantial than the previous ones as the intention is also to raise problems and provoke discussion.

In the first section we calculate the steady state inventories of ^{90}Sr and ^{137}Cs in the Danish terrestrial environment; we assume a constant annual fallout rate of 1 mCi km^{-2} of each of the two radionuclides. We further assess the transfers of ^{90}Sr and ^{137}Cs between important parts of the terrestrial ecosystem.

The next section discusses the toxicity concept; it appears that toxicity of a substance is not an absolute but depends essentially on the context in which the toxic substance is considered.

Some practical applications of radioecological sensitivity and variability are shown in connection with siting evaluation and in relation to remedial measures in case of a major environmental radioactive contamination. Finally in the conclusion further environmental radioactivity studies are proposed.

5.2. Inventories and transfers of ^{90}Sr and ^{137}Cs in the Danish terrestrial ecosystem

"Tror I, det er hele Verden!" sagde Moderen; den strækker sig langt på den anden Side Haven, lige ind i Præstens Mark; men det har jeg aldrig været!

DEN GRIMME ELLING

"Do you suppose this is the whole world?" said their mother. "Why, it goes a long way past the other side of the garden, right into the parson's field; but I've never been as far as that.

THE UGLY DUCLING

An inherent difficulty when dealing with ecosystems is its confinement. All ecosystems have some connection with the surrounding world. In this respect the Danish terrestrial ecosystem is no exception. However, in theory agricultural production in Denmark is sufficient for the Danish food and fodder supply.

The aim of this section is for steady state theory to estimate the inventories and annual transfer rates of ^{90}Sr and ^{137}Cs in the terrestrial ecosystem, which just covers Denmark. For the purpose the ecosystem is divided into four major parts: the soil, the vegetable agricultural products, the animal agricultural products and the human population.

5.2.1. Definitions

The "soil" comprises the total Danish abiotic terrestrial environment; furthermore it includes all living organisms not dealt with under agricultural products in the present context. We shall thus not consider transport and inventories of radionuclides in wild flora and fauna, nor in microorganisms. Although such a study may be essential it is not pertinent to the present context where our main objective is the important sources to the human diet of ^{90}Sr and ^{137}Cs. The "soil" also comprises those parts of the cultivated crops which neither are grazed nor harvested but which remain in the fields. The dried weight of the upper 20 cm of the Danish soil - the ploughing layer - is estimated as the mean weight of the layer at the ten Danish experimental farms: 2.55×10^{8} kg km^{-2} (RRD 75).

The "vegetable agricultural products" are the vegetable products grazed or harvested at the 3×10^{4} km^{2} used for agricultural purpose in Denmark.

The "animal agricultural products" are composed of the stocks of cattle, pigs and poultry and of their produce. Other domestic animals e.g. sheep and horses are neglected because they are very few in comparison to the above mentioned categories.

We assume the Danish "human population" at equilibrium to be five million and the death rate (and birth rate) to equal 65,000 per year. We shall neglect immigration as well as emigration.

5.2.2. Calculations

To accomplish the calculations of inventories and transfer rates of ^{90}Sr and ^{137}Cs in the Danish ecosystem we suppose a constant fallout rate of 1 mCi km^{-2} y^{-1} of the radionuclide considered, and we assume the fallout distributed throughout the year to be the global ^{90}Sr fallout (cf. fig. 1.3.1). At steady state the situation has persisted so long that the annual total removal from radioactive decay, run off, etc. equals the annual supply of activity from fallout. If the mean residence time of the radionuclide considered in the Danish terrestrial ecosystem is T_m years the equilibrium level is T_m mCi km^{-2}.

According to D.1.4.2. the ^{90}Sr run-off in Denmark is 9.3% of the total deposit. For ^{137}Cs we estimate the run-off as 1/5 of the ^{90}Sr run-off (Ya63, Voi71), i.e. to 1.8 % of the total ^{137}Cs deposit. Wind erosion is neglected because the Danish soil is not wind eroded to any large extent; furthermore the main part of the wind-eroded soil remains in the terrestrial ecosystem.

In case of ^{90}Sr the annual removal rate due to run-off is 0.093 mCi km^{-2} at steady state and the radioactive decay is consequently 1-0.093 = 0.907 mCi km^{-2} corresponding to a terrestrial steady state deposit of $0.907\ (1- \exp(-\ln 2/28))^{-1}$ = 37.1 mCi ^{90}Sr km^{-2}. In the case of ^{137}Cs the steady state level becomes $0.982\ (1- \exp(-\ln 2/30))^{-1}$ = 43 mCi ^{137}Cs km^{-2} assuming a total ^{137}Cs run-off of 1.8 %. In this calculation we have neglected export and import of activity from agricultural and fishery products to the system, because these sources - as we shall see in the following - are immaterial in this context.

The production of Danish vegetable agricultural produce in 1975 is given in table D.5.2.1. in the appendix. We have assumed that 1975 represented a steady state situation. The standing crop of grass during summertime, which was used as reference time of the year for the calculations of the inventories, was estimated to be 9 Tg (~ 1 kg grass m^{-2} (RRD63, RRD77).

Table 5.2.2.1. The annual steady state production and consumption levels of ^{90}Sr and ^{137}Cs in Curies from Danish vegetable agricultural produce for an annual deposition rate of 1 mCi km^{-2} y^{-1} (of ^{90}Sr or ^{137}Cs respectively)

Product	Production		Fodder		Human diet	
	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
Grain etc.	0.18	0.21	0.18	0.20	0.0043	0.0079
Straw	1.38	0.33	0.43	0.104	-	-
Roots	0.168	0.0099	0.14	0.008	0.0010	0.0014
Tops	0.44	0.046	0.38	0.039	-	-
Grass etc.	1.26	0.54	1.10	0.47	-	-
Vegetables	0.0016	0.0003	-	-	0.0021	0.0004
Fruits	0.0002	0.0006	-	-	0.0004	0.0010
Total	3.43	1.14	2.23	0.82	0.0078	0.0107

It was assumed that there was neither import nor export of vegetable agricultural products. The grain actually exported was supposed used as fodder instead of imported concentrates. Keeping this in mind, the figures were calculated from the data in table D.5.2.1. and from the radioecological sensitivities calculated in appendix C. In the calculation of "Grain etc." the last column of table 2.2.2.1. was applied together with table C.2.3.1. for flour and grits. Straw, root crops and grass were estimated from tables C.2.4.3. (Nos. 4 and 5), C.2.4.4. (No. 4), C.2.4.1. (Nos. 1, 2 and 3) and C.2.4.2. (Nos. 7 and 8) respectively. Tables C.2.5.1. and C.2.5.3. were applied for vegetables and fruits.

Table 5.2.2.1 shows the production and division of ^{90}Sr and ^{137}Cs in Danish vegetable agricultural products. The inventories at midyear are equal to the production figures except for grass which contains 9/23 of the production levels because the weight ratio between standing crop and annual production of grass is 9:23. Hence the inventories in vegetable agricultural products become 2.7 Ci ^{90}Sr and 0.81 Ci ^{137}Cs.

The Danish livestock in 1975 consisted of 3.1×10^6 cattle, 7.7×10^6 pigs and 13.4×10^6 hens; the living weights - estimated from the numbers and weights of the various age groups (Da 57-77, Da 68) - were 1 Tg, 0.5 Tg and 0.02 Tg respectively. The annual production of animal agricultural produce appears in the appendix in tables D.5.2.2 and D.5.2.3 (milk products).

Table 5.2.2.2. The annual steady state production, consumption and export levels of ^{90}Sr and ^{137}Cs in millicuries from Danish animal agricultural produce for an annual deposition rate of 1 mCi km^{-2} y^{-1} (of ^{90}Sr or ^{137}Cs respecticely)

	Production		Fodder		Human diet		Export	
	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
Cattle	116	14.9	(76)	(2.5)	0.12	2.5	35	4.6
Pigs	145	36	(49)	(3.1)	0.19	7.4	82	21
Poultry + eggs	7.6[a]	0.9[b]	-	-	0.01	0.34	2.4	0.43
Bone and meat meal	117	5.6	125	5.6	-	-	-	-
Whole milk, cream	2.8	3.9	0.49	0.69	2.2	3.1	0.13	0.17
Skimmed milk etc.	6.2	8.7	5.3	7.5	0.75	1.0	-	-
Dried milk	4.8	6.8	0.94	1.31	-	-	3.9	5.5
Whey	1.33	7.2	1.33	7.2	-	-	-	-
Cheese	4.1	0.62	-	-	1.36	0.21	2.7	0.41
Butter	0.07	0.10	-	-	0.03	0.03	0.03	0.07
All milk products Σ	19.3	27	8.1	16.7	4.3	4.3	6.8	6.2
Total	288	79	133	22	4.7	14.5	126	32

a) 3 mCi ^{90}Sr were from eggs and 4.6 mCi from poultry.

b) 0.1 mCi ^{137}Cs were from eggs and 0.8 mCi from poultry.
The figures in the table were calculated from the production figures in tables D.5.2.2. and D.5.2.3. and from the radioecological sensitivities in appendix C. Tables C.3.4.1. and C.3.4.2. were applied for beef and pork (recalling that 1 kg meat contains 0.1 g Ca and 3.2 g K). As the observed ratio between pCi ^{90}Sr $(g\ Ca)^{-1}$ in poultry bone and diet equals the ratio between eggs and diet of the hen (Co66a), the radioecological sensitivity of poultry equaled that of eggs (= 2.5 pCi ^{90}Sr $(g\ Ca)^{-1}$ y per mCi ^{90}Sr km^{-2} cf. table C.3.6.1). The radioecological sensitivity of poultry was for ^{137}Cs estimated at 3.5 times that of eggs (cf. 3.6.2), i.e. of 6.3 pCi ^{137}Cs kg^{-1} y per mCi ^{137}Cs km^{-2} (cf. table C.3.6.2) or 2 pCi ^{137}Cs $(g\ K)^{-1}$ y per mCi ^{137}Cs km^{-2}. The radioecological sensitivities used for the various milk products were from tables C.3.2.1. and C.3.2.2.

The production and division of ^{90}Sr and ^{137}Cs in Danish animal agriculture are shown in table 5.2.2.2. The inventories in the livestock become 0.31 Ci ^{90}Sr and 0.049 Ci ^{137}Cs.

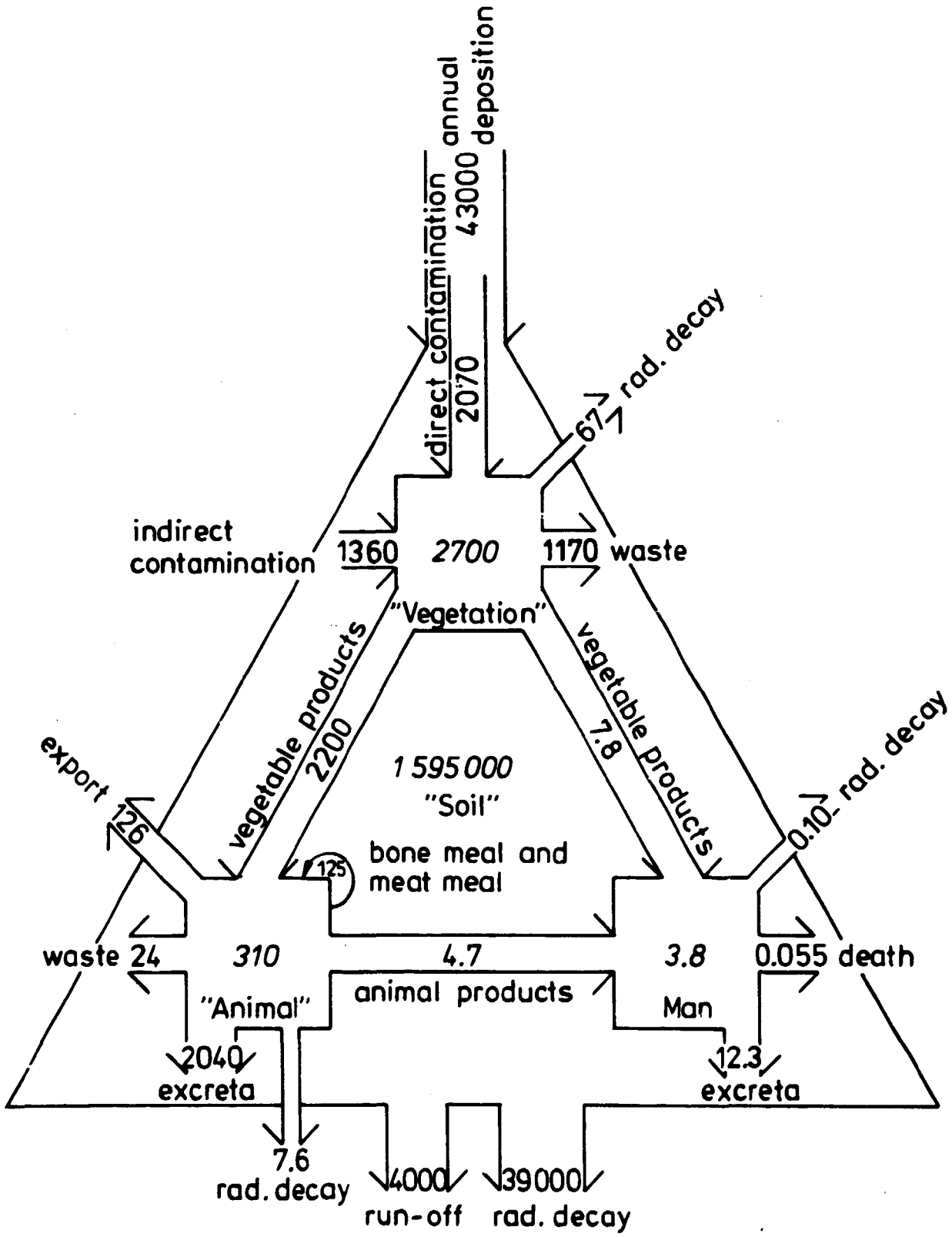

Fig. 5.2.3.1. Inventories (italics) and annual transfers of ^{90}Sr in the Danish terrestrial environment (43 000 km^2), assuming steady state at an annual deposition rate of 1 mCi ^{90}Sr km^{-2} y^{-1}. All figures are mCi ^{90}Sr.

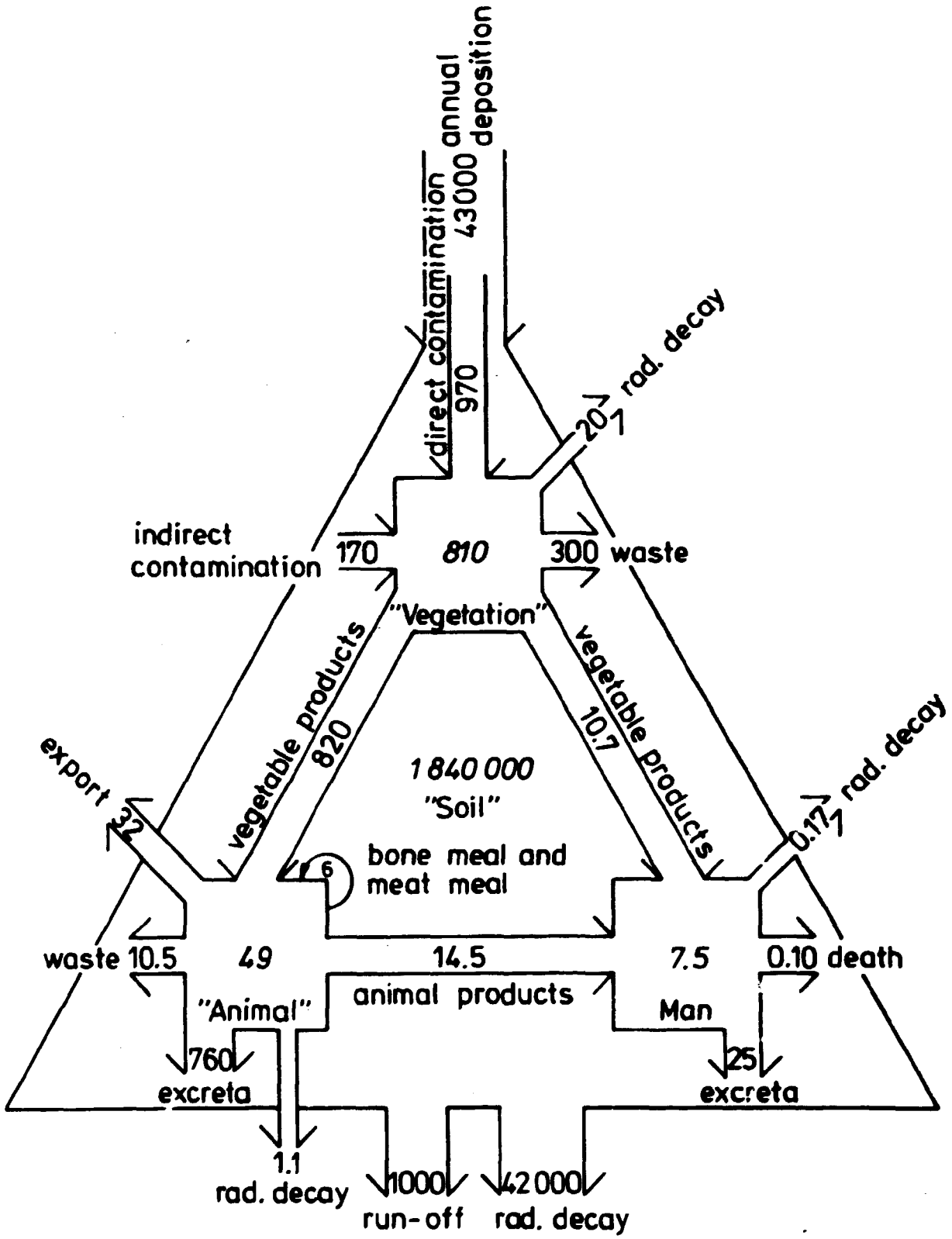

Fig. 5.2.3.2. Inventories (italics) and annual transfers of ^{137}Cs in the Danish terrestrial environment (43 000 km^2) assuming steady state at an annual deposition rate of 1 mCi ^{137}Cs km^{-2} y^{-1}. All figures are mCi ^{137}Cs.

The age-weighted weight of the Danish human population is 0.3 Tg. The potassium content is 0.6 Gg assuming 2 gK kg^{-1} living weight (RRD59-76) and the Ca content is 4.6 Gg, calculated from the Ca content of the various age groups as estimated by MITCHELL et. al. (Mi45). Assuming the mean radioecological sensitivities in humans to be equal to 0.82 pCi ^{90}Sr $(gCa)^{-1}$ y per mCi ^{90}Sr km^{-2} (4.3.2) and to 2.85 x 4.4 = 12.5 pCi ^{137}Cs $(gK)^{-1}$ y per mCi ^{137}Cs km^{-2} (4.5.2 and 4.2.2), the inventories of the Danish population become 3.8 mCi ^{90}Sr and 7.5 mCi ^{137}Cs at steady state for an annual fallout rate of 1 mCi km^{-2} of each of the two radionuclides.

5.2.3. Discussion of the models

The circulation of ^{90}Sr and ^{137}Cs in the Danish terrestrial ecosystem at steady state is shown in figs. 5.2.3.1 and 5.2.3.2 respectively. Table 5.2.3 summarizes the relative inventories and concentrations of ^{90}Sr and ^{137}Cs together with those of the two chemical congeners: Ca and K.

It has sometimes been discussed whether ^{90}Sr or ^{137}Cs show biomagnification in the food chain. It is evident from table 5.2.3., that the concentration of ^{90}Sr is approx. three times higher in animals than in the vegetable fodder. In the case of ^{137}Cs we notice an increase in the animals by approx. 60%. However in humans the ^{90}Sr concentration is 4 times lower than in vegetation and 14 times lower than in domestic animals. The ^{137}Cs concentration in humans is between the concentrations found in vegetable and animal products. As compared to Ca the ^{90}Sr level in humans is approx. 80 times lower than in vegetable products and 13 times lower than in animals. The $^{137}Cs/K$ ratio on the other hand shows a tendency to increase along the human food chain. We may conclude that herbivores are more subceptible to contamination with ^{90}Sr than omnivores (and carnivores), whereas in ^{137}Cs the situation is reversed. The ^{137}Cs concentrations are lower in omnivorous man than in herbivorous lifestock, because the vegetable fodder of the animals show higher ^{137}Cs concentrations than the human vegetable diet.

The pyramid of numbers of ^{90}Sr is different from that of Ca indicating that nature discriminates against Sr relative to Ca. The potassium pyramid is similar to that of the ^{137}Cs pyramid.

Table 5.2.3. Pyramids of numbers for the inventories of Ca, ^{90}Sr, K and ^{137}Cs in the Danish terrestrial ecosystem

	Relative units				$^{90}Sr/Ca$	$^{137}Cs/K$
	Ca	^{90}Sr	K	^{137}Cs		
Soil: Ploughing layer 0-20 cm	10^6 (1)	10^6 (1)	10^6 (1)	10^6 (1)	1	1
Vegetation: Standing crops (cf. table D.5.2.1 and 5.2.2)	1.4×10^3 (0.4)	1.7×10^3 (0.5)	1.1×10^3 (0.3)	4×10^2 (0.12)	1.2	0.4
Animals: Lifestock (cf. table D.5.2.2 and 5.2.2)	0.9×10^3 (6)	2.0×10^2 (1.4)	3×10^1 (0.24)	3×10^1 (0.19)	0.2	0.8
Man:	1.6×10^2 (6)	2.4×10^0 (0.1)	4×10^0 (0.14)	4×10^0 (0.15)	0.015	1.1

The Ca content of the 0-20 cm ploughing layer of the Danish soil was 30 Tg (2.7 g Ca kg^{-1} (cf. 4.2.3) and the K content was 161 Tg (14.7 g K kg^{-1} (RRD75)). The calcium and potassium concentrations in the various products were estimated from appendix C.

The Ca, ^{90}Sr, K and ^{137}Cs levels in the table were relative to those in soil, which were fixed at 10^6.

The figures in brackets were the relative concentrations.

In figs 5.2.3.1 and 5.2.3.2 the divisions between direct and indirect contamination of vegetable products were estimated from the pertinent prediction models in Appendix C. In case of ^{90}Sr 60% came from direct and 40% from indirect contamination; for ^{137}Cs the figures were 85% and 15% respectively. For ^{90}Sr the root uptake from the soil compare to the amounts returning to the soil as unused vegetable material, or in other words the ^{90}Sr transferred from vegetable products to animal products and to humans nearly equals the direct contamination of ^{90}Sr.

The transfer of ^{90}Sr from vegetable products to animal produce is nearly 3 times higher than the transfer of ^{137}Cs due to the higher radioecological sensitivity of crops to ^{90}Sr than to ^{137}Cs (table 5.2.2.1). From animal products to humans however, the ^{137}Cs transfer is 3 times the ^{90}Sr transfer because the bones of the lifestock retain most of the ^{90}Sr.

The transfer of ^{137}Cs from vegetable products to humans is also higher that of ^{90}Sr, because grain products (white bread) as well as potatoes show higher radioecological sensitivity to ^{137}Cs than to ^{90}Sr. The total transfer of ^{137}Cs to humans is twice that of ^{90}Sr. This is a little higher than the ratio found in table 4.2.2, because the diet composition applied in the present estimate was that from 1975, whereas table 4.2.2 used the 1962 composition (cf. D.4.2.1).

With the export of animal agricultural products 126 mCi ^{90}Sr and 32 mCi ^{137}Cs are leaving the ecosystem; these amounts are 0.3% and 0.07% of the annual input of the two radionuclides and immaterial as compared to radioactive decay and run-off to the sea.

The net uptake of ^{90}Sr and ^{137}Cs in the animal agricultural system was,respectively, 12.4% and 7.8% of the inputs to the systems; in humans the corresponding figures were 1.2% and 1.1%. The turnover rates in the animal agricultural system are thus approx. ten times higher than in the human system for ^{90}Sr and seven times higher for ^{137}Cs. The production rate of the animal system was 1.05 x lifestock per year while the human "production" was only 0.013 x population per year. Hence the relative productivity of the animal sytem was nearly 80 times that of the human system, which explains the lower turnover rate of radionuclides in the latter system.

In the models for the Danish terrestrial ecosystem we have neglected input from the marine environment, because it is difficult to delimit the marine ecosystem. However, for the total human diet the contribution of ^{90}Sr from fish is nil and for ^{137}Cs it is in the order of 2-3%. The animal agriculture also receives marine products through its use of fish meal; however compared to the vegetable fodder the contributions of ^{90}Sr and ^{137}Cs from fish meal were less than 1%.

We have also neglected the drinking water in our system. The drinking water supply to the Danish livestock is approx. 10^{11} l y^{-1}. The annual contribution of ^{90}Sr will be between 0.7-30 mCi y^{-1}, depending upon whether the water is ground water or stream water (table C.1.4.1),i.e. probably less than 1% of the

intake from vegetable products. For ^{137}Cs the contribution will be even lower (1.4).

If all vegetable products for human consumption were imported from a contamination-free area the ^{90}Sr intake by humans would be reduced by a factor of 2.7 and the ^{137}Cs intake by a factor of 1.7. The effective dose equivalent commitment (D.4.7.7) from diet intake of ^{90}Sr and ^{137}Cs could be reduced by a factor of two. The same reduction could be obtained if instead the animal agricultural products for Danish human consumption were imported. But animal products are much more expensive than vegetable products.

5.3. Physiological and environmental radiotoxicity

havde Dyrene ikke været giftige og kyssede af Hexen, da vare de blevne forvandlede til røde Roser, men Blomster blev de dog, ved at hvile på hendes Hoved og ved hendes Hjerte;

DE VILDE SVANER

If the creatures hadn't been poisonous and kissed by the witch, they would have been turned into red roses; though, mind you, they did change into flowers, just from resting on her head and at her heart.

THE WILD SWANS

Exposure of humans to radiation may lead to the incidence of stochastic and non-stochastic health effects. Non-stochastic effects occur only if certain levels of dose are exceeded; these levels are several orders of magnitude in excess of doses typically received from radioactive contamination of the environment under normal peaceful conditions. The important stochastic effects are carcinogenesis in the exposed population and hereditary effects in its progeny. The probability of occurrence of stochastic effects is assumed proportional to dose, without threshold.

The toxicity of a substance or agent may be given by the so called $LD_{50(30)}$ value, expressing the dose (e.g. in grams or Joules pr. kg living weight of the exposed organism) which within 30 days kills 50% of a population exposed to the dose. $LD_{50(30)}$ may be applied for non-stochastic effects of ionizing radiation, but for stochastic effects the effective doseequivalent commitment is preferable (cf. D.4.7.7).

The physiological radiotoxicity deals with the toxicity of radionuclides in the human diet ingested and in the air breathed by humans.

The environmental radiotccicity is about the toxicity of radionuclides deposited on the Earth's surface (cf. the UNSCEAR model in 1.1).

5.3.1. Physiological radiotoxicity

We shall define the physiological radiotoxicity of a given substance as the per caput effective dose-equivalent commitment to an adult "standard man" received from the intake of one unit of radioactivity of the substance. This intake could either be peroral or by inhalation. As we have seen (1.7.2) the relative toxicity of radionuclides depends upon the route of intake. Strontium-90 is thus more hazardous than ^{239,240}Pu if the intake is peroral, while plutonium becomes the more toxic if inhalation is the route of entry.

The physiological radiotoxicity from oral intake of a radionuclide is:

$$K \times P_{34} \times P_{45} \times w_T \qquad \text{(Eq 5.3.1)}$$

where K is a constant which convertes the diet intake to the prober unit of activity

P_{34} is the transfer factor from diet to tissue of the radionuclide

P_{45} is the transfer factor from activity content in tissue to dose to tissue of the radionuclide

w_T is ICRP's weighting factor (Ic77) for the tissue T.

If the radionuclide enters more than one tissue type the various contributions to the whole-body dose should be added in order to calculate the physiological radiotocicity of the radionuclide.

In case of inhalation the expression becomes:

$$P_{14} \times P_{45} \times w_T \qquad \text{(Eq 5.3.2)}$$

where P_{14} is the transfer factor from atmosphere to tissue. Analogous with peroral intake the dose contributions to various tissues should be added in order to calculate the toxicity.

Let us calculate the physiological toxicity of ^{90}Sr and ^{137}Cs in the Danish diet. In this calculation we will use Bq as the unit of activity and assume intakes of 1 Bq of each of the two radionuclides. We will calculate the effective dose equivalent commitment from these intakes and consider the results as expressions for the physiological toxicity (as to transfer factors and weighting factors cf. 4.7.7):

^{90}Sr: $0.0433[0.16 \cdot 1.4 \cdot 0.12 + 0.16 \cdot 1.9 \cdot 0.03] \cdot 10^4 =$ $15.6\ nSv(Bq)^{-1}$

^{137}Cs: $0.0211 \cdot 2.85 \cdot 1.8 \cdot 10^{-2} \cdot 10^4 = 10.8\ nSv(Bq)^{-1}$

The transfer factors for ^{90}Sr are based upon the pCi $^{90}Sr\ (gCa)^{-1}$ ratios and for ^{137}Cs on the pCi $^{137}Cs(gK)^{-1}$ ratios.

The annual intakes with Danish diet of Ca and K is 623 g and 1281 g respectively (table D.4.2.1.a).

One Becquerel equals 27 pCi. Hence the K factor for ^{90}Sr becomes $\frac{27}{623} = 0.0433$ pCi Bq^{-1} y $(gCa)^{-1}$ and for ^{137}Cs: $\frac{27}{1281} = 0.0211$ pCi $\cdot$ $Bq^{-1} \cdot y\ (gK)^{-1}$.

The calculation shows that the physiological radiotoxicity of ^{90}Sr in the Danish diet is 1.44 times that of ^{137}Cs. If the Faroese diet is considered the Ca and K amounts are 415 g and 1174 g, respectively (Table 4.2.4). The physiological toxicities t on increase to $\frac{623}{415} \cdot 15.6 = 23\ nSv\ (Bq)^{-1}$ for ^{90}Sr and to $\frac{1281}{1174} \cdot 10.8 = 11.8\ nSv\ (Bq)^{-1}$ for ^{137}Cs

The physiological radiotoxicity may thus be influenced by other substances in the diet that the radionuclide itself, as seen above, e.g. by the chemical congeners to the radionuclides.

5.3.2. Environmental radiotoxicity

Recurring to the environmental radiotoxicity it becomes even more evident that this quantity depends upon the interaction of numerous factors, where the physiological toxicity of the radionuclide is only one - and not necessarily the most important.

In analogy with the physiological radiotocicity the per caput environmental radiotoxicity is calculated as:

$$q \times P_{23} \times P_{34} \times P_{45} \times w_T \qquad \text{(Eq 5.3.3)}$$

where q is a constant which converts the deposition of activity to the proper unit for the toxicity calculation
P_{23} is the transfer factor from Earth surface to human diet, in other words the radioecological sensitivity of human diet for the given radionuclide in the given environment.

In 4.7.7 the effective dose equivalent commitment has been calculated for depositions of 73 mCi ^{90}Sr km^{-2} and of 117 mCi ^{137}Cs km^{-2}. In Denmark the dose from ^{90}Sr became 10.08 + 3.42 = 13.5 mrem and for ^{137}Cs (from diet) 26 mrem. If we normalize to 1 Bq m^{-2} of each of the two radionuclides we find the per caput environmental radiotoxicity to be

$$\frac{13.5 \cdot 10^4 \cdot 27}{73000} = 50 \text{ nSv } (\text{Bqm}^{-2})^{-1} \text{ for } ^{90}Sr$$

and

$$\frac{26 \cdot 10^4 \cdot 27}{117000} = 60 \text{ nSv } (\text{Bqm}^{-2})^{-1} \text{ for } ^{137}Cs,$$

i.e. the environmental radiotoxicity of ^{137}Cs in Denmark is 20 per cent higher than that of ^{90}Sr. If we calculate the radiotoxicities of the two radionuclides for the Faroese environment we find 53 $nSv(Bqm^{-2})^{-1}$ for ^{90}Sr and 181 $nSv(Bqm^{-2})^{-1}$ for ^{137}Cs, i.e. in the Faroes Islands ^{137}Cs is 3.4 times more toxic than ^{90}Sr. In the Greenland environment ^{137}Cs is twice as toxic as ^{90}Sr.

We have seen that the per caput environmental radiotoxicity depends upon the radioecological sensitivity (P_{23} in Eq. 5.3.3). The collective environmental radiotoxicity furthermore depends upon the diet yield per unit area. Although the radioecological sensitivity of e.g. ^{137}Cs in Faroese milk thus is ten times higher than in Danish milk the collective dose from milk per unit area in the Faroe Islands is lower than the collective dose of milk produced per unit area in Denmark because the Danish milk production per unit area is nearly fifty times higher than the Faroese.

In order to calculate the collective environmental radiotoxicity the per caput radiotoxicity is multiplied by the number of people which may be supplied by the food production in a unit area of the environment considered. Denmark is self-supplying with human diet and we furthermore export milk and meat-products. We may estimate the exportation of ^{90}Sr and ^{137}Cs for human consumption from the information given in Tables D.5.2.2, D.5.2.3 and 4.2.2. We find that Danish export of ^{90}Sr is 50% of our consumption while the export of ^{137}Cs is twice our consumption. In other words 1 km^2 of Denmark supply

$$\frac{5 \cdot 10^6 \cdot 1.5}{43000} = 174 \text{ people with } "^{90}Sr \text{ diet}"$$

and

$$\frac{5 \cdot 10^6 \cdot 2}{43000} = 233 \text{ people with } "^{137}Cs \text{ diet}"$$

We may now calculate the collective environmental radiotoxicity of ^{90}Sr and ^{137}Cs of 1 km^2 of Denmark:

$$^{90}Sr: 174 \cdot 50 \cdot 10^{-3} = 8.7 \text{ µman Sv } (Bqm^{-2})^{-1} km^{-2}$$

and

$$^{137}Cs: 233 \cdot 60 \cdot 10^{-3} = 14 \text{ µman Sv } (Bqm^{-2}) km^{-2}$$

In the Faroe Islands 68% of the ^{90}Sr and 94% of the ^{137}Cs is of local origin (cf. table 4.2.4).

Hence the Faroese environment can supply

$$\frac{0.68 \cdot 4 \quad 10^4}{1400} = 19.4 \text{ people km}^{-2} \text{ with "}^{90}\text{Sr diet"}$$

and

$$\frac{0.94 \cdot 4 \cdot 10^4}{1400} = 27 \text{ people km}^{-2} \text{ with "}^{137}\text{Cs diet"}$$

The collective environmental radiotoxicity of 1 km^{-2} of the Faroe Islands becomes

$$53 \cdot 19.4 \cdot 10^{-3} = 1.03 \text{ µman Sv } (Bqm^{-2})^{-1} \text{ km}^{-2} \text{ for } ^{90}\text{Sr}$$

and

$$27 \cdot 181 \cdot 10^{-3} = 4.9 \text{ µman Sv } (Bqm^{-2})^{-1} \text{ km}^{-2} \text{ for } ^{137}\text{Cs}$$

The environmental radiotoxicity of the Faroese environment is thus lower than of the Danish environment if collective doses of ^{90}Sr and ^{137}Cs are considered, while the toxicity was higher in the Faroe Islands with regard to per caput doses.

5.3.3. Comparison of radiotoxicities

Table 5.3.3. summarizes the various diet-derived radiotoxicities of ^{90}Sr and ^{137}Cs in Denmark and the Faroe Island. It should be noted that the units of the figures in the three rows differ; but the table demonstrates that toxicity is not an absolute, but strongly dependent upon the context in which we use the concept. For a given contamination an inhabitant in the Faroes will show higher individual doses than a Dane exposed to the same contamination. The collective dose however will be considerably higher in the Danish environment due to the higher food production per unit area in Denmark. The environmental toxicity of ^{90}Sr is lower than that of ^{137}Cs and the difference between the two radionuclides is most pronounced for the collective doses.

The calculations of the collective radiotoxicities in Table 5.3.3. are based on the average foodproduction per unit area for the total areas of Denmark and the Faroe Islands, which is

Table 5.3.3. Radiotoxicities of ^{90}Sr and ^{137}Cs in Denmark and the Faroe Islands

Radiotoxicity		Denmark		Faroe Islands	
		^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
Physiological: nSv per Bq ingested		15.6	10.8	23	11.8
Environmental	Per caput nSv per Bq m^{-2} deposited	50	60	53	181
	Collective μmanSv km^{-2} per Bq m^{-2} deposited	8.7	14	1.03	4.9

the pertinent approach in case of global fallout. However, if the contamination has a local nature, as e.g. in the case of the releases from a nuclear installation, the results may change. If, e.g., pure agricultural areas in the Faroe Islands and in Denmark are compared the collective radiotoxicities is not necessarily still higher in Denmark, because the lower productivity of Faroese agricultural areas may be more than compensated for by the higher radioecological sensitivities of the Faroese agricultural products.

We have seen that a high radioecological sensitivity is not necessarily tantamount to a high collective environmental radiotoxicity. There may even be a tendency to an inverse relationship between the radioecological sensitivity and the collective environmental radiotoxicity, because a high radioecological sensitivity often is found in agricultural areas with low productivity (Cham 70, Aa72a). In Jutland, where the mean yield of wheat in a wheatfield is 485 tons km^{-2} (Da57-77) the radioecological sensitivity is 1.33 times higher than in a wheatfield in the Islands, where the yield is 519 tons km^{-2}, whereas the ratio between the collective environmental radiotoxicities is only 1.24.

The present definition of radiotoxicity infers that only fatal cancers (apart from those of the skin) and the hereditary effect in the first two generations are included in the health detriment (cf. D.4.7.7). If the total radiotoxicity of a radioactive substance should be evaluated one might furthermore include fatal cancers of the skin, non-fatal cancers and subsequent hereditary effects. This has not been done because a common measure of the health detriment for these various effects is not available. The total radiotoxicity of a substance as ^{131}I, which may give rise to non-fatal cancers of the thyroid is thus underestimated as long as it is based on effective dose-equivalent commitment only, and the same would be the case for ^{85}Kr, which irradiates the skin.

Although the effective dose-equivalent commitment may thus not be a perfect description of the health detriment and hence of the toxicity of a radioactive substance, we are, regarding the definition of toxicity, in a far better position for radioactive substances than for most other toxic materials.

5.4. Applications

"Snik snak!" sagde Soldaten, "vil du strax sige mig, hvad du vil med det, eller jeg trækker min Sabel ud og hugger dit Hoved af!"
FYRTØJET

"Rubbish! said the soldier. "Tell me at once what you want to do with it - or I'll have out my sword and cut your head off".
THE TINDER-BOX

In the evaluation of the suitability of a potential nuclear site numerous factors are taken into account. One of these - but not necessarily the most weighty - are the factors which influence the dose to humans from ingestion of radionuclides released during the operation of the nuclear plant. These doses may, as we have discussed in the preceeding section, be individual or collective; both types should be considered.

5.4.1. Other radionuclides compared with ^{90}Sr and ^{137}Cs

The present study has concentrated on ^{90}Sr and ^{137}Cs. Together these nuclides have delivered nearly 90% of the internal effective dose equivalent to humans as the result of ingestion of food contaminated with the fallout radionuclides from nuclear weapons testing.

We have seen that the environmental behaviour of the two nuclides differs markedly. In the Danish environment ^{90}Sr may thus represent a radionuclide entering the foodchain by indirect as well as direct contamination, whereas ^{137}Cs may be characteristic for a contaminant that preferentially is incorporated through direct contamination. Most radionuclides will be like ^{137}Cs in this respect.

We have furthermore seen that the direct contamination of crops with various radionuclides strongly depends upon their ability to translocate within the plants. Cesium-137 is a nuclide which is highly translocated (VIII), and thus likely to be relatively more abundant in seeds and fruits than most other radionuclides.

Throughout the study fallout radionuclides other than ^{90}Sr and ^{137}Cs have also been considered. In grain we have thus examined ^{54}Mn, ^{89}Sr and $^{239,240}Pu$. The transfer factors of these radionuclides from deposition to cereals did not exceed those of ^{137}Cs (2.2.3, 2.2.4 and IX) and we have found that the local variabilities of ^{54}Mn, ^{89}Sr and $^{239,240}Pu$ were within the same range as the corresponding variabilities of ^{90}Sr and ^{137}Cs (cf. Table B.2.2.9).

In a recent study of lead in Danish cereal grain (So 78) we observed that stable Pb also shows a local variability in Danish grain which is within the range of the various fallout radionuclides studied.

In the case of milk ^{89}Sr and ^{131}I have been determined in countrywide collected samples when possible. Neither of these radionuclides have shown higher transfer factors than those of ^{90}Sr and ^{137}Cs (3.2.3). The local variability of ^{89}Sr in milk was between that of ^{90}Sr and ^{137}Cs. In the case of ^{131}I the

local variability was determined from a countrywide sampling performed in the last 3 weeks of September 1962 (RRD62) of fresh milk (Fig. A.1.4.2.1), dried milk (Fig. A.1.3.2) and untreated milk (Fig. A.1.1.3.1). From this study the local variability of ^{131}I in Danish milk was estimated at 0.3-0.4, i.e. similar to that of ^{137}Cs in milk (Table B.3.2.3).

5.4.2. A hypothesis

Several radionuclides may be released from nuclear installations under normal operations as well as in case of accidents. Some of these radionuclides have hitherto not been present in quantities such that studies of their radioecological behaviour in various environments have been feasible. However, it is possible to evaluate a terrestrial environment radioecologically by applying the radioecological sensitivities and variabilities of ^{90}Sr and ^{137}Cs only.

In the environmental terrestrial studies of other radionuclides we have found no indications of transfer factors higher than those found for ^{90}Sr and ^{137}Cs. We have not seen examples either of local variabilities exceeding those observed for these two radionuclides, and the local pattern in Denmark of the various radionuclides - and of lead, too - follows that of ^{90}Sr and ^{137}Cs. The levels in the western part of the country is at most 2-3 times higher than those in eastern Denmark. The radioecological sensitivities of agricultural products from Jutland are seldom more than two times those from the Islands; the mean ratios between the two halves of the country for grain, vegetables and milk are 1.6 for ^{90}Sr and 1.3 for ^{137}Cs.

We may therefore conclude, that from a terrestrial radioecological point of view Jutland is less suited than the Islands for nuclear installations, because the individual doses through the terrestrial foodchains on the average are higher than those received from the same releases in the Islands.

In the preceeding section (5.3) we discussed the importance of the agricultural production at the site. This production would determine the collective dose along with the radicecological sensitivity. The yields per unit area of arable land are usual-

ly 5-10% higher in the Islands than in Jutland*). Hence the difference between collective doses at the two sites will be less than the difference between individual doses, but Jutland will still be the most sensitive area to radioactive contamination of terrestrial foodchains.

However, nuclear installations also release radionuclides to the marine environment. In this case it is not sufficient to consider radioecological sensitivities of ^{90}Sr and ^{137}Cs only. Neither of these radionuclides show especially high concentration factors in marine organisms (2.7.2, 3.5.2 and 3.6). Whereas many activation products released with waste water from nuclear reactors, e.g. ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{60}Co and ^{65}Zn may show a substantial biomagnification in marine organisms, e.g. in brown algae and in mussels (RRD77). Also fission products (^{95}Zr, ^{106}Ru) and transuranic elements ($^{239,240}Pu$, ^{241}Am) may show concentrations in the marine biota which are orders of magnitude higher than those in the surrounding seawater.

For some radionuclides e.g. radiocesium low salinities are tantamount to higher biomagnification than high salinities. As the waters around Jutland on the average show higher mean salinities than the waters around the Islands, the radioecological sensitivity of the marine environment in the western part of the country may be lower than that of the eastern, assuming the effective mean life of a given radionuclide to be the same in the two environments. The per caput doses from consumption of marine products would thus be higher in the Islands than in Jutland. The catch of fish around the Islands is, however, an order of magnitude less than the catch in the waters around Jutland. Hence the collective dose from radionuclides with waterborne releases may be higher for a nuclear plant in Jutland than for a similar plant in the Islands. UNSCEAR (Un77) has calculated that for present days nuclear reactors approx. 3.5% of the total effective dose equivalent commitment come from the terrestrial foodchains, and approx. 4% from water path ways.

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*) In Jutland 68% of the total area are arable area of farms in the Islands 66% are arable.

5.4.3. Considerations in case of nuclear accidents

During the normal operation of a nuclear power reactor only minor releases of radionuclides take place. For light water reactors (BWR and PWR) the airborne releases mostly consist of noble gases such as ^{133}Xe, ^{85}Kr and of ^{131}I. The waterborne releases are the activation products mentioned above (5.4.2) and radiocesium. The doses through the radioecological pathways are generally an order of magnitude lower than the doses from inhalation and from external radiation from the air or from the ground (Un77). Hence for dose assessments the population densities around nuclear sites are usually more important than the agricultural production.

In the case of a very serious accident, where the fuel elements in the reactor core melt and where the reactor containment is ruptured, substantial amounts of ^{90}Sr and especially of ^{137}Cs may be released to the environment along with many other radionuclides, principally radicactive noble gasses and radioiodine. The long term doses and the contamination of land from such an accident will largely depend upon the amounts of the longlived and medium-longlived radionuclides released from the accident. Strontium-90 and ^{137}Cs are the most important in this group of radionuclides.

The time of the year influences the doses from the foodchain (cf. D.4.7.8). In Denmark ^{137}Cs is not taken up by the roots to any large extent. If the accident happens during the winter the diet will therefore contain relative low concentrations of ^{137}Cs as compared to a summer accident. In the case of ^{90}Sr the difference will be less pronounced because the indirect contamination of crops with ^{90}Sr is of the same order as the direct. In any event the ^{137}Cs levels will decrease more rapidly than the ^{90}Sr levels. The doses from the food could be substantially reduced by discarding the first year's harvest.

To elucidate this a little further the radioecological sensitivity may be divided into an instant and a delayed sensitivity. The instant radioecological sensitivity is the contribution to the total radioecolcgical sensitivity which, according to the

prediction models, is delivered within the same year as the contamination has occurred; the delayed radioecological sensitivity is the remainder.

As a high time variability among years ($CV_{p(years)}$) is tantamount to primary dependence on direct (instant) fallout, it is obvious to express instant radioecological sensitivity as a function of the annual variability as shown in Fig. 5.4.3.

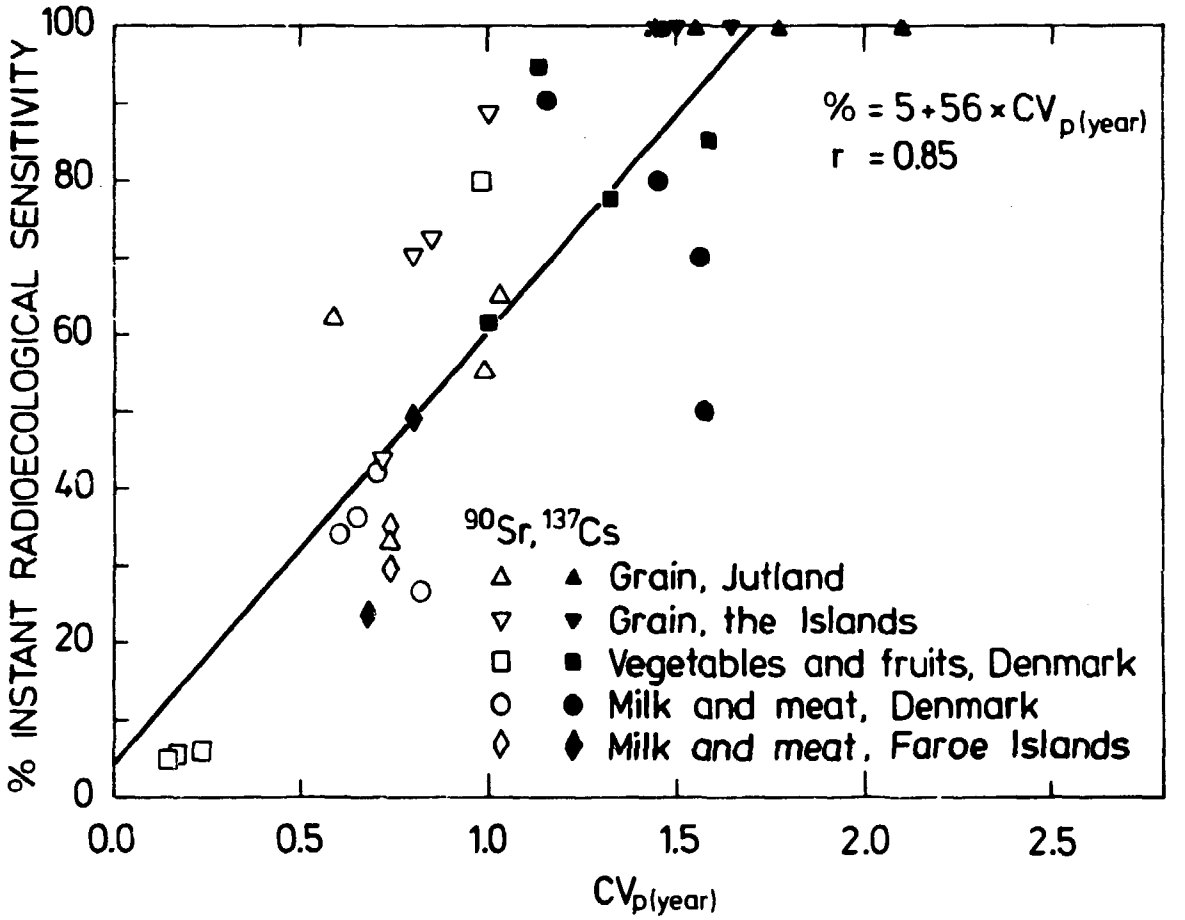

Fig. 5.4.3. The instant radioecological sensitivity (cf. the text) as a function of the timevariability ($CV_{p(years)}$) determined 1962-19 '. The tables in appendix B were used for the varabilities and appendix C delivered the prediction models for the calculation of the instant radioecological sensitivities.

The equation in Fig. 5.4.3, establish connection between variability and radioecological sensitivity. From this equation, it is possible after an accident to estimate the effect of discarding the first years harvest of a given crop grown in the area contaminated by the accident. Let e.g. the crop be rye

from Jutland. In that case $CV_{p(years)} = 1$ for ^{90}Sr (table B.2.2.7) and the dose commitment from ^{90}Sr in rye would then be reduced to approx 40% of the dose received if no remedial measures had been taken (cf. also D.4.7.8).

If the equation in Fig. 5.4.3 is applied to animal agricultural products one should keep in mind that the fodder to the animals delivering the products should be from the same year*) as the products themselves. As this is only approximately the case the observed instant radioecological sensitivity for the animal produce in fig. 5.4.3 is generally lower than that calculated from the time variabilities of the samples.

Fig. 5.4.3 shows that a substantial reduction in the doses from ^{137}Cs in Danish diet components could be obtained by discarding the first years harvest. In the case of ^{90}Sr the effect is more modest. It would, e.g., have nearly no effect on the ^{90}Sr doses if the first years harvest of vegetables (cabbage, carrots and potatoes) were discarded. In the Faroe Islands the effect of discarding first years harvest on ^{90}Sr as well as on ^{137}Cs doses would generally be less than in Denmark.

The longtime dose from an accident will however only to minor degree come from the foodchain. The main contributor to the dose will be the external radiation from the ^{137}Cs deposited.

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*) In case of milk and meat the products from $May_{(i)}$ to $April_{(i+1)}$ should be from fodder produced in the $year_{(i)}$ (cf. 3.2.1 and 3.4.1).

5.5. Conclusions

5.5.1. General

The public concern of pollution in general and of radioactivity in particular has made radioactive contamination of the environment a controversial issue. It is not easy to inform the population in order to improve the understanding of these often very complicated problems. The task is even harder because miscomprehensions are by no means limited to laymen's circles. The treatment of the toxicity concept in public debate is an excellent example of present misunderstandings of pollution problems. A large proportion of the public is reluctant to accept nuclear energy as one way of energy production among several others; to a considerable degree this may be due to three inherent properties of ionizing radiation: 1) It cannot be detected directly with any of the human senses, 2) The equipment used to measure radioactivity is extremely sensitive, which means that even very low - and from a health point of view insignificant - amounts are measurable, and 3) It is - contrary to most other pollutants - relatively easy to quantify the health detriment of a given dose of ionizing radiation to the population.

In this final chapter we have especially dealt with the application of radioecological sensitivity and variability in the evaluation of health detriment from environmental radioactive contamination. We have furthermore demonstrated how the radioecological sensitivity can be applied for the calculation of inventories and transfers of radionuclides in an ecosystem.

5.5.2. Inventories and transfers

The annual transfers of ^{90}Sr and ^{137}Cs from vegetable agricultural produce to animal produce are 80% and 100%, respectively, of the inventories of the standing crop at summer. From animals to man the corresponding figures are 1.5% and 30% of the animal inventories, and from vegetable produce to humans 0.3% and 1.3% of the vegetable inventories.

The inventories of ^{90}Sr were 3 times that of ^{137}Cs for "vegetation", 6 times for "animals" and 0.5 times for humans, assuming steady state and equal fallout rates of the two radionuclides.

The producers are thus in general more sensitive to ^{90}Sr than to ^{137}Cs contamination, and so are the consumers if they are herbivorous. For humans as omnivores sensitivity to environmental ^{137}Cs contamination is greater than that of a ^{90}Sr contamination.

The agricultural statistic of the Faroe Islands is less comprehensive than the Danish statistics, and significant amounts of human diet and animal fodder are imported to the Faroe Islands. Hence it is not possible to calculate detailed models for the Faroese ecosystem as we have done for the Danish. However, we may estimate that the ^{90}Sr inventories of Faroese vegetation are half the ^{137}Cs inventories, and that the domestic animals in the Faroe Islands contain approx. 3 times more ^{90}Sr than ^{137}Cs for the same environmental contamination of the two nuclides.

The ^{137}Cs inventory of the Faroese population is estimated to be approx. 6 times the ^{90}Sr inventory. This shows that the Faroese terrestrical ecosystem behaves differently from the Danish with regard to ^{90}Sr and ^{137}Cs contamination.

5.5.3. Radiotoxicities

We have considered physiological and environmental radiotoxicity. We have shown that the first mentioned for ^{90}Sr in Denmark is 50% higher than that of ^{137}Cs. In the Faroes the physiological toxicity of ^{90}Sr is two times that of ^{137}Cs, if the two radionuclides are ingested with Faroese diet. The environmental radiotoxicity of ^{137}Cs is higher than that of ^{90}Sr. In the Faroes the per caput environmental radiotoxicity of ^{137}Cs is 3 times that in Denmark but the collective environmental radiotoxicity of ^{137}Cs is 3 times higher in Denmark than in the Faroes due to the higher agricultural production pr unit area in Denmark.

The examples show that it is confusing or even misleading to state that one substance is more toxic than another without specifying the circumstances under which the two toxicities are compared.

5.5.4. Applications

The radioecological sensitivities of ^{90}Sr and ^{137}Cs can be used to classify an environment radioecologically. If the terrestrial environmental samples from one environment show higher radioecological sensitivities for both radionuclides than the samples from another environment, our conclusion would be that the former environment may result into higher individual doses from ingestion of local food than the latter, also for releases of radionuclides other than ^{90}Sr and ^{137}Cs.

As the collective doses pr unit area are proportional to the foodproduction pr unit area and as the radioecological sensitivity often is inverse proportional to the crop yield, high individual doses in an area are not necessarily tantamount to high collective doses.

The so-called instant radioecological sensitivity may be related to the time variability observed for ^{90}Sr and ^{137}Cs in the various environmental samples collected since the beginning at the sixties. The two quantities are proportional and the time variability of a sample may thus be used for the estimation of the instant radioecological sensitivity, which again is a relative measure of the dose received within the same year as the activity release. The time variabilities may thus be applied to estimate the effect of discarding the first years harvest or of changing the agricultural production in a contaminated area.

5.5.5. Further studies

Because of its many relations to nearly all diciplines of science an environmental study will inevitably inspire further studies. First of all it raises a number of basic problems which should be dealt with in well-defined experimental studies. This final section will, however, emphasize further environ-

mental studies, which the present ones are related to.

We have in Appendix C shortly mentioned dynamic modelling of the environmental behaviour of radionuclides. Such models have until now been of limited usefulness because the basic data, e.g., on transfer coefficients between the various compartments, often have been poor. The present study has estimated a number of transfer coefficient for ^{90}Sr and ^{137}Cs in the Danish environment. It may be worthwhile to apply these data in a dynamic model for the behaviour of these radionuclides in the Danish environment. Sensitivity analyses are an important application of dynamic models. By such analyses one may identify which data are of special importance and thus where future research is waranted.

Although ^{90}Sr and ^{137}Cs have been studied comprehensively in the terrestrial environment for the last 20 years there still exist some uncertainty in assessing the long term transfer of these radionuclides to humans and their diet; especially because the migration down through the soil and any physical or biochemical processes, which modify the availability of the radionuclides for uptake into plants with time, have not been finely settled. Hence it is important still to follow the behaviour of ^{90}Sr and ^{137}Cs in soil as well as in terrestrial produce and in humans.

In the very long term perspective other radionuclides might dominate the environmental contamination arising from present day's use of nuclear fission. Technetium-99 with a half-life of 2.14×10^5 years, ^{129}I with 1.57×10^7 years half-life and the transuranic elements (e.g. $^{239,240}Pu$ and ^{241}Am) are such nuclides. Our understanding of the environmental behaviour of the transuranic elements in these years are rapidly improving, whereas radioecological studies of ^{99}Tc and ^{129}I have only just begun. None of these two radionuclides are especially hazardous, but they seem to be transferred quite easily through foodchains and research efforts are warranted for a better understanding of their long-term behaviour in the environment.

Our knowledge of the behaviour of radionuclides in the terres-

trial environment is generally better than that of the marine environment, especially the oceans. We can point out problems such as the relatively high marine levels along the Greenland eastcoast, and the future fate of the 0.8 MCi ^{137}Cs hitherto (1979) released to the sea from the reprocessing plant of Windscale. The behaviour of radionuclides released into the deep sea, e.g. from waste disposal, will also require further studies in the coming years. Marine studies are expensive and international cooperation is therefore a must for this research.

With a rapidly increasing world population other methods of foodproduction than those at present in use are foreseen. Sea vegetation may thus be an important part of the future diet of the world. This should be taken into account in marine radioecological studies, because sea plants show high concentration factors for many environmental pollutants. Generally, in future radioecological studies one should be vigilant to new path-ways to humans.

A special application of the environmental contamination with radionuclides is the use of the nuclides for identification and dating purposes. Lake sediments may thus be age-determined from their contents of ^{137}Cs or $^{239,240}Pu$. From ^{90}Sr analysis of, e.g., a bone sample one may make out whether the sample originates from before "the nuclear age". However, one should be careful not to jump to conclusions. We have thus recently seen that a human bone sample, which had been laying in a spruce forest for 10 years contained considerly more ^{90}Sr than expected from its age, presumably because the bone material had accumulated ^{90}Sr from the surroundings. Isotope ratios are often useful for identification of sources of contamination. Studies of $^{238}Pu/^{239,240}Pu$ ratios or of $^{134}Cs/^{137}Cs$ ratios may thus show whether a given contamination arises from nuclear power installations or from weapons testing. Measurements of such ratios do not require quantitative analyses where the chemical yields are known because we assume that isotopes of the same element found in the environment at some distance from the sources are of the same chemical form.

Although our knowledge of radioactive contamination of the en-

vironment in many respects is incomplete, it is nevertheless considerably more comprehensive than our knowledge of non-radioactive pollutants. Primarily because the sensitivity to detection and assessment of radioactive substances is generally far superior to that of other pollutants. The many possible sources of non-radioactive pollutants (local, regional and global) combined with no easy method to determine the age of a non-radioactive substance contribute to the difficulties in assessing pathways to man. Reliable transfer factors between the various environmental compartments are thus not easily available. It should however be possible to improve the situation by using more systematic and comprehensive sampling programs, e.g. similar to those used for radioactive contamination in the present study. By analysis of variance one could then get a better understanding of the various mechanisms determining the non-radioactive environmental contamination. As an example of such a procedure we have recently carried out a study of the lead contamination of Danish cereal grain collected since 1962 at the Danish State Experimental Farms (So78). We shall in future studies include other previous collected environmental samples, and we intend also to examine other metals than lead. We expect that such a study, among other things, could show whether the concept of radioecological sensitivity and variability also could be applied to non-radioactive pollutants.

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The quotations heading the various sections in the main text are from the original Danish text of HANS CHRISTIAN ANDERSEN's Fairy Tales translated into English by R.P. KEIGWIN.

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DANSK RESUME

Det foreliggende arbejde har haft til hensigt at kvantificere de radioøkologiske egenskaber af miljøprøver. Til dette formål er benyttet "den radioøkologiske sensitivitet" og "variabiliteten". Den førstnævnte størrelse er identisk med overføringsfaktoren fra deponeret radioaktivt globalt nedfald til den pågældende miljøprøve, medens variabiliteten er den såkaldte partielle variationskoefficient beregnet på grundlag af en variansanalyse.

De atmosfæriske kærnevåbenforsøg har siden sprængningen af det første nukleare våben i 1945 kontamineret omgivelserne med en række forskellige radionukleider. Til de længere levende stoffer blandt disse hører ^{90}Sr og ^{137}Cs, der på grund af deres kemiske slægtskab med henholdsvis calcium og kalium forholdsvis let overføres fra det abiotiske miljø til levende organismer. Målingerne af ^{90}Sr og ^{137}Cs koncentrationerne i luft, vand og jord, som er beskrevet i kapitel 1, har dannet basis for beregningen af overføringsfaktorerne fra det radioaktive nedfald via fødekæden frem til mennesket, og de abiotiske miljøprøvers variabilitet er benyttet ved vurderingen af de biologiske prøvers kontaminationsveje.

Miljøfaktorer som klima, jordbundsforhold og landbrugspraksis påvirker, som det omtales i kapitel 2, den radioøkologiske sensitivitet såvel som variabiliteten af radionukleid koncentrationerne i vegetabilske prøver. Afgrøder med et stort overflade til vægtforhold og en langsom vækst udviser almindeligvis større direkte kontamination end planter med de modsvarende egenskaber. For Danmarks vedkommende spiller kornprodukterne en særlig rolle, fordi rugbrød, hvori hele kærnen anvendes, indgår som en væsentlig bestanddel af kosten. Den direkte kontamination af kornet er i dette tilfælde af speciel betydning, hvorfor der er udført en række eksperimentelle kontaminationsforsøg af kornplanter til belysning af forholdene. Indirekte kontamination (rodoptagelse af radioaktive stoffer) er en vigtig transportvej for specielt ^{90}Sr til f.eks. grøntsager. På Færøerne er jordbundsforholdene årsag til at også ^{137}Cs optages i betydelig grad via rødderne, hvilket resulterer i en høj radioøkologisk sensitivitet af færøske afgrøder med hensyn

til ^{137}Cs. Lav indtager en særstilling på grund af sin exceptionelt høje radioøkologiske sensitivitet.

Kapitel 3 omhandler den radioaktive forurening af dyrene og deres produkter først og fremmest komælken. Et højt indhold af græs i kvægfoderet resulterer i højere ^{137}Cs niveauer i mælken, end hvis foderet fortrinsvis er baseret på roer; derimod er ^{90}Sr niveauerne i mælken kun lidt påvirkelige af en sådan ændring i fodersammensætningen. Den høje indirekte ^{137}Cs kontamination af det færøske græs betyder, at den radioøkologiske sensitivitet af færøsk mælk med hensyn til ^{137}Cs er væsentlig større end af dansk mælk. De terrestriske dyr, som indgår i den humane fødekæde udgøres primært af herbivorer, hvorfor niveauerne i kødet er afhængige af plantefoderets radioaktivitetsindhold; de extreme niveauer, som findes i lav afspejles således i en høj radioøkologisk sensitivitet af rensdyrkød. De marine dyr i fødekæden er hovedsageligt carnivorer; men selv om der sker en opkoncentrering af et nukleid som ^{137}Cs gennem de marine fødekæder giver dette sig ikke udslag i specielt høje niveauer i marine dyr, da radioaktivitetskoncentrationerne i havet er relativt lave og fordi havvandets forholdsvis høje saltkoncentrationer begrænser ^{137}Cs optagelsen i havets organismer. Den radioøkologiske sensitivitet med hensyn til ^{137}Cs kan under danske forhold for visse marine fisk nærme sig sensitiviteten af danske husdyr, dels fordi radioaktivitetsniveauerne i danske farvande aftager relativt langsomt, og dels fordi danske husdyr hovedsageligt kun modtager ^{137}Cs via direkte kontamination af vegetationen.

I det fjerde kapitel omtales den radioaktive forurening af mennesket, dets kost og af modermælk. De vegetabilske produkter i menneskets kost er almindeligvis de vigtigste ^{90}Sr kilder, mens ^{137}Cs primært kommer fra de animalske produkter. Dog er mælk og kornprodukter uanset deres oprindelse begge vigtige kilder til såvel ^{90}Sr som ^{137}Cs i kosten. I dansk kost stammer således ca. 70 % af kostens ^{90}Sr og ca. 55 % af dens ^{137}Cs fra disse to kostgrupper. Færøsk og grønlandsk kost er karakteriseret ved deres relativt høje radioøkologiske sensitivitet med hensyn til ^{137}Cs, hovedsageligt stammende fra de lokale terrestiske animalske produkter. Den danske, færøske og grønlandske

kost udviser næsten den samme radioøkologiske sensitivitet med hensyn til ^{90}Sr. De effektive dosis ekvivalent commitmenter fra radioaktivt nedfald fra kærnevåbenforsøg er beregnet til 1.6 mSv for en dansker, 4.2 mSv for en færing og 1.6 mSv for en grønlænder. Cæsium-137 bidrog med godt og vel halvdelen til disse dosisekvivalenter. Den højere dosis til færinger skyldes den større nedbørsmængde og dermed større nedfaldshastighed på Færøerne; men det skyldes tillige den større radioøkologiske sensitivitet af det færøske miljø.

Det samlede maksimale antal alvorlige sundhedsskader i den danske, færøske og grønlandske befolkning, som følge af den radioaktive forurening fra hidtidige atmosfæriske kærnevåbenforsøg, skønnes at ligge i størrelsesordenen et hundrede fatale cancertilfælde og et tilsvarende antal alvorlige genetiske skader.

I femte og sidste kapitel vises hvorledes resultaterne i det foregående kan benyttes til at opstille en model for ^{90}Sr og ^{137}Cs omsætningen i det danske terrestriske økosystem under ligevægtsbetingelser. Begrebet radiotoxicitet defineres, og der opstilles kvantitative udtryk for en fysiologisk og en miljøbetinget radiotoxicitet. Det vises, at et stofs (radio)toxicitet er et relativt begreb, som afhænger af den sammenhæng, hvori begrebet anvendes. Endelig demonstreres, at radioøkologisk sensitivitet kan benyttes ved bedømmelsen af et miljøs egnethed som recipient for radioaktive udslip, samt at virkningen af modforholdsregler i form af kassation af afgrøder eller omlægning af landbrugsproduktionen kan estimeres udfra afgrødernes tidsvariabilitet.

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